

# Catalyst for polymerization of olefins and process for polymerization of olefins using the same.

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## Abstract

A catalyst for the polymerization of olefins comprising (A) a solid catalyst component essentially containing magnesium, titanium, an electron donor compound, and a halogen which is prepared by contacting a magnesium compound, a titanium halide compound, and an electron donor compound, (B) an organoaluminum compound, and (C) an organosilicon compound and a process for polymerizing an olefin using the same are disclosed, the organosilicon compound (C) being represented by formula (I): wherein  $R<1>$  and  $R<2>$ , which may be the same or different, each represents an alkyl group having from 1 to 3 carbon atoms; and the cyclohexyl group and the cyclopentyl group may be substituted. Using the catalyst, a polyolefin having high stereoregularity and broad molecular weight distribution can be obtained in high yield.



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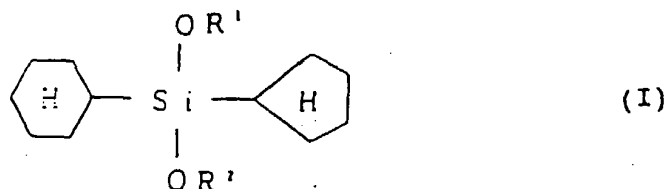
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**Catalyst for polymerization of olefins and process for polymerization of olefins using the same.**

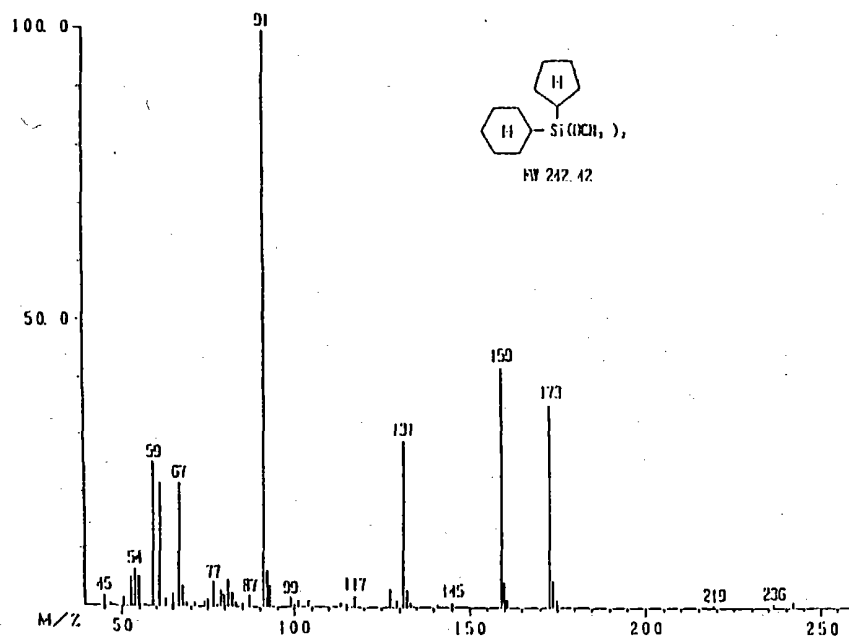
A catalyst for the polymerization of olefins comprising (A) a solid catalyst component essentially containing magnesium, titanium, an electron donor compound, and a halogen which is prepared by contacting a magnesium compound, a titanium halide compound, and an electron donor compound, (B) an organoaluminum compound, and (C) an organosilicon compound and a process for polymerizing an olefin using the same are disclosed, the organosilicon compound (C) being represented by formula (I):



wherein R<sup>1</sup> and R<sup>2</sup>, which may be the same or different, each represents an alkyl group having from 1 to 3 carbon atoms; and the cyclohexyl group and the cyclopentyl group may be substituted. Using the catalyst, a

polyolefin having high stereoregularity and broad molecular weight distribution can be obtained in high yield.

FIG. 1



## FIELD OF THE INVENTION

This invention relates to a catalyst for the polymerization of olefins and a process for the polymerization of olefins using the same. More particularly, it relates to a catalyst for the polymerization of olefins with which an olefin polymer having high stereoregularity and broad molecular weight distribution can be obtained in high yield and a process for polymerizing an olefin in the presence of the catalyst.

## BACKGROUND OF THE INVENTION

Hitherto, a large number of specific organosilicon compounds for use as an electron donor (external electron donor) as a component of a Ziegler-Natta catalyst or for use as an electron donor (internal electron donor) contained in a solid catalyst component of a Ziegler-Natta catalyst have been proposed for the purpose of producing polymers having improved stereoregularity or enhancing catalytic activity in olefin polymerization using the catalyst.

Various proposals have been made on processes for producing this kind of organosilicon compounds. For example, U.S. Patent 4,977,291 proposes a process for producing a silicon compound having at least one cycloalkyl group in which a silicon compound containing an aromatic group as a starting compound is hydrogenated in the presence of a catalyst, e.g., a Raney nickel catalyst.

U.S. Patent 4,958,041 discloses a process for producing a diorganodialkoxysilane having at least one branched alkyl group other than the two alkoxy groups in which a tetraalkoxysilane or a monoorganotrialkoxysilane is reacted with a Grignard reagent having the structural formula  $\text{RMgX}$  wherein R is an alkyl group or a cycloalkyl group and X is a halogen atom.

In JP-A-5-255350 is disclosed a cycloalkoxysilane represented by the formula  $(\text{R}'\text{O})_x(\text{R}')_y\text{Si}(\text{OR})_{4-x-y}$  for use as an electron donor component of a Ziegler-Natta catalyst for olefin polymerization, wherein each R is independently selected from alkyl groups having 1 to 5 carbon atoms and acyl groups having 2 to 5 carbon atoms, each R' is independently selected from a cyclopentyl group, a cyclohexyl group, a cycloheptyl group, and substituted groups thereof, x is 1, 2, 3, or 4, and y is 0, 1, or 2. The term "JP-A" as used herein means an "unexamined published Japanese patent application." JP-A-5-310757 discloses tert-butoxycyclopentyl-diethoxysilane as a novel silane compound and a process for producing the same.

For homo- or copolymerization of olefins a number of processes using a catalyst system comprising a solid catalyst component essentially containing magnesium, titanium, an electron donor compound, and a halogen, an organoaluminum compound, and an organosilicon compound have been proposed.

For example, JP-A-57-63310 and JP-A-57-63311 propose a process for polymerizing an olefin having 3 or more carbon atoms using a catalyst system comprising (a) a solid catalyst component containing a magnesium compound, a titanium compound and an internal electron donor, (b) an organoaluminum compound, and (c) an organosilicon compound having an Si-O-C bond. However, these processes are not always satisfactory for obtaining highly stereoregular polymers in high yield, and further improvement has been demanded.

On the other hand, JP-A-63-3010 discloses a catalyst system for the polymerization of olefins and a process for polymerizing olefins using the same, the catalyst system comprising (a) a solid catalyst component prepared by bringing a dialkoxymagnesium, an diester of aromatic dicarboxylic acid, an aromatic hydrocarbon, and a titanium halide into contact and subjecting the resulting product in a powdered state to a heat treatment, (b) an organoaluminum compound, and (c) an organosilicon compound.

JP-A-1-315406 discloses a catalyst system for olefin polymerization and a process for polymerizing an olefin using the same, the catalyst system comprising (a) a solid catalyst component prepared by bringing titanium tetrachloride into contact with a suspension of diethoxymagnesium in an alkylbenzene, adding phthalic acid dichloride thereto to react to obtain a solid product, and further contacting the resulting solid product with titanium tetrachloride in the presence of an alkylbenzene, (b) an organoaluminum compound, and (c) an organosilicon compound.

JP-A-2-84404 proposes a catalyst system for the polymerization of olefins and a process for homo- or copolymerizing an olefin(s) using the same, the catalyst system comprising (a) a solid titanium catalyst component essentially containing magnesium, titanium and a halogen which is prepared by bringing a magnesium compound and a titanium compound into contact, (b) an organoaluminum compound catalyst component, and (c) an organosilicon compound catalyst component containing a cyclopentyl group or a derivative thereof, a cyclopentenyl group or a derivative thereof, or a cyclopentadienyl group or a derivative thereof.

Each of these known techniques aims at such high catalytic activity that a step of removing residual catalyst components, such as chlorine and titanium, from the resulting polymer (a so-called de-ashing step)

may be omitted and, at the same time, an improvement in yield of a stereoregular polymer or an improvement in durability of the catalytic activity for polymerization, and has achieved excellent results to their purpose.

In recent years, however, it has been pointed out that the olefin polymers obtained by polymerization using these catalyst systems comprising such a highly active catalyst component, an organoaluminum compound and an organosilicon compound have narrower molecular weight distribution as compared with those obtained by using conventional catalyst systems comprising a titanium trichloride type catalyst component in combination with an organoaluminum compound and, if desired, an electron donor compound as a third component. For polyolefins to have narrower molecular weight distribution means poorer moldability, leading to less applicability.

Various manipulations have been suggested to solve this problem. For example, adoption of a multi-stage polymerization system has been proposed for obtaining polyolefins with broader molecular weight distribution. Nevertheless, a multi-stage polymerization system requires repetition of tedious and complicated operation of polymerization and also involves a step for recovery of a chelating agent to be used for polymerization and is not therefore deemed to be favorable from the considerations of labor and cost.

As the latest technique, JP-A-3-7703 discloses a process for polymerizing an olefin in the presence of a catalyst system comprising (a) a solid titanium catalyst component essentially containing magnesium, titanium, a halogen, and an internal electron donor, (b) an organoaluminum compound, and (c) at least two organosilicon compounds as an external electron donor. According to this process, a polyolefin having broad molecular weight distribution can be obtained without involving laborious multi-stage polymerization operation. However, the use of at least two organosilicon compounds as an electron donor for polymerization makes the process still tedious and complicated.

Further, solid catalyst components containing an aluminium halide, a magnesium compound and a titanium halide as essential components, and catalysts containing such a solid catalyst component, an organic aluminium compound and as the third component, an organic acid ester or a silicone compound are known. For example, JP-A-55-161807 discloses a catalyst comprising (i) a composition obtained by pulverizing magnesium chloride, an organic acid ester, a halogenated hydrocarbon and an aluminium halide together and then carrying out heat treatment with titanium tetrachloride, (ii) an organic aluminium compound, and (iii) an organic acid ester; and JP-A-61-31402 discloses a catalyst comprising (i) a solid catalyst component obtained by reacting a reaction product of an aluminium halide and a silicone compound with a magnesium compound and then reacting with a titanium halide and an ester of phthalic acid, (ii) an organic aluminium compound and (iii) a silicone compound.

Furthermore, solid catalyst components containing an alkoxyaluminium compound, a magnesium compound and a titanium halide as essential components, and catalysts for polymerization of olefins which contain such a solid catalyst component, an organic aluminium compound and as the third component, an organic acid ester or a silicone compound are also known. For example, JP-A-57-145104 discloses a catalyst component obtained by pulverizing magnesium chloride, an organic acid ester and an alkoxyaluminium compound together and then carrying out heat treatment with titanium tetrachloride; and JP-A-1-245002 discloses a catalyst comprising (i) a solid catalyst component obtained by bringing diethoxymagnesium into contact with titanium tetrachloride, followed by addition of an trialkoxyaluminium and then reacting with phthalic dichloride, (ii) an organic aluminium compound, and (iii) an epoxy-p-menthane compound.

These solid catalyst components and catalysts as described above have been developed as a result of studies for attaining high catalytic activity in polymerization of propylene so that the amount of the solid catalyst component can be minimized and the step of removing a catalyst residue (e.g., chlorine and titanium) remaining in a resulting polymer can be omitted, for improving the yield of a stereoregular polymer, or for improving durability of the catalytic activity for polymerization, and they provide good results for the respective objects. However, none of these catalysts can produce stereoregular polymers having a density of from 0.900 to 0.906 g/ml without lowering the yield of stereoregular polymers insoluble in a polymerization solvent as used in polymerization of olefins, particularly polymerization of propylene, according to the slurry method.

In the case where olefins, particularly propylene are polymerized by the slurry method in the presence of the aforesaid highly active catalyst, the resulting polymer has a high stereoregularity and is obtained in a high yield as compared with the case where a catalyst comprising a conventional titanium trichloride type solid catalyst component, an organic aluminium compound and an electron donor compound is used. However, the density of the resulting polymer tends to be higher than 0.906 g/ml, giving rise to various problems, e.g., breaking in rapid rolling rate and deterioration in transparency of a film product molded by a BOPP molding.

It has been known that the density of the resulting polymer can be controlled to some extent by lowering the polymerization temperature or introducing a small amount of ethylene as a comonomer into the polymerization system of olefins (particularly propylene) in the presence of the aforesaid highly active catalyst. In case of the slurry method, however, an undesirable phenomenon occurs that a low molecular weight polymer which is soluble in a polymerization solvent is formed at a high rate, and in the polymerization of propylene or copolymerization of propylene and ethylene, an atactic polypropylene which has an extremely poor stereoregularity is generated at a high rate. The atacticity can be evaluated in terms of the content of soluble portions of the resulting polymer or copolymer in a polymerization solvent, which is hereafter referred to "RDS" (the content of reactor diluent solubles).

Increase of the RDS in the slurry polymerization gives rise to problems with respect to the production cost of a polymer and the stability in operation because a reactor and a pipeline are stained, and an extraction step is required after separation of the particles of a resulting polymer from a polymerization solvent. Further, generation of fine powders contained in the resulting polymer, particularly those having a particle size of 100 micron or less, tend to cause clogging in a pipeline in the polymerization process, and other problems in the step of separation and drying of the polymer.

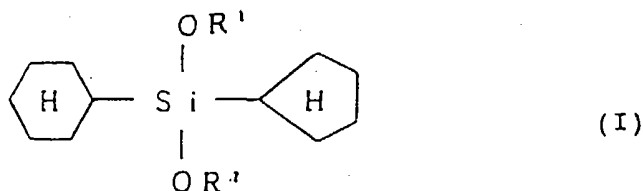
#### SUMMARY OF THE INVENTION

An object of the present invention is to provide a catalyst for the polymerization of olefins which can eliminate the above-mentioned problems associated with the conventional techniques, i.e., a catalyst system which exhibits high polymerization activity through simpler operation and provides an olefin polymer having broad molecular weight distribution while maintaining a satisfactory yield of a highly stereoregular polymer.

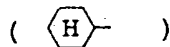
Another object of the present invention is to provide a process for polymerizing an olefin to produce a polyolefin having broad molecular weight distribution and high stereoregularity in high yield.

As a result of extensive investigations, the present inventors have found that an olefin polymer having high stereoregularity and broad molecular weight distribution can be obtained in high yield by polymerizing an olefin(s) in the presence of a catalyst comprising (A) a solid catalyst component essentially containing magnesium, titanium, an electron donor compound, and a halogen, which is formed by bringing a magnesium compound, a titanium halide compound, and an internal electron donor compound into contact, (B) an organoaluminum compound, and (C) an organosilicon compound having a specific structure. The present invention has been completed based on this finding.

The present invention provides a catalyst for the polymerization of olefins comprising (A) a solid catalyst component containing magnesium, titanium, an internal electron donor compound, and a halogen, which is prepared by bringing a magnesium compound, a titanium halide compound, and an internal electron donor compound into contact, (B) an organoaluminum compound, and (C) an organosilicon compound represented by formula (I):



wherein  $\text{R}^1$  and  $\text{R}^2$ , which may be the same or different, each represents an alkyl group having from 1 to 3 carbon atoms; and the cyclohexyl



group and the cyclopentyl



5 group may be substituted.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a chart showing the results of MS with which cyclohexylcyclopentylmethoxysilane was identified.

Fig. 2 is a chart showing the results of two-dimensional analysis by <sup>1</sup>H-NMR/<sup>13</sup>C-NMR (COSY spectrum) with which cyclohexylcyclopentylmethoxysilane was identified.

Fig. 3 is a chart showing the results of IR with which cyclohexylcyclopentylmethoxysilane was identified.

#### DETAILED DESCRIPTION OF THE INVENTION

Organosilicon compound (C) which can be used in the present invention is an asymmetric organosilicon compound having a cyclohexyl group or a derivative thereof and a cyclopentyl group or a derivative thereof both directly bonded to the silicon atom. The cyclohexyl group and the cyclopentyl group may be substituted with an alkyl group preferably having 1 to 3 carbon atoms or a halogen atom. The number of substituent on each ring is preferably not more than two.

Examples of the alkyl group for R<sup>1</sup> and R<sup>2</sup> in formula (I) include methyl, ethyl, n-propyl, and isopropyl. Of these, methyl and ethyl are preferred.

The organosilicon compound of the present invention, which is represented by formula (I) described above, is a cyclohexylcyclopentylalkoxysilane. Examples of the cyclohexylcyclopentylalkoxysilane include cyclohexylcyclopentylmethoxysilane, cyclohexylcyclopentylethoxysilane, cyclohexylcyclopentyl-n-propoxysilane, and cyclohexylcyclopentylisopropoxysilane. Of these, cyclohexylcyclopentylmethoxysilane and cyclohexylcyclopentylethoxysilane are preferred organosilicon compounds for use as an electron donor serving as a component of an olefin polymerization catalyst.

Various derivatives of these asymmetric organosilicon compounds are included within the scope of formula (I). In particular, those having one or two substituents, such as a methyl group, chlorine or bromine, at the 3-, 4- or 5-position of the cyclohexyl group thereof and/or one or two substituents as exemplified above at the 2-, 3- or 5-position of the cyclopentyl group thereof are preferred. Two substituents may be at the same position of the cyclohexyl or cyclopentyl group. Plurality of the substituents on the same ring may be the same or different.

Specific examples of the derivatives of the asymmetric organosilicon compounds are 3-methylcyclohexylcyclopentylmethoxysilane, 3-methylcyclohexylcyclopentylethoxysilane, 3-methylcyclohexylcyclopentyl-n-propoxysilane, 4-methylcyclohexylcyclopentylmethoxysilane, 4-methylcyclohexylcyclopentylethoxysilane, 4-methylcyclohexylcyclopentyl-n-propoxysilane, 3,5-dimethylcyclohexylcyclopentylmethoxysilane, 3,5-dimethylcyclohexylcyclopentylethoxysilane, 3,5-dimethylcyclohexylcyclopentyl-n-propoxysilane, 3,3-dimethylcyclohexylcyclopentylmethoxysilane, 4,4-dimethylcyclohexylcyclopentylmethoxysilane, cyclohexyl-2-methylcyclopentylmethoxysilane, cyclohexyl-2-methylcyclopentylethoxysilane, cyclohexyl-2-methylcyclopentyl-n-propoxysilane, 3-methylcyclohexyl-2-methylcyclopentylmethoxysilane, 3-methylcyclohexyl-2-methylcyclopentylethoxysilane, 3-methylcyclohexyl-2-methylcyclopentyl-n-propoxysilane, 4-methylcyclohexyl-2-methylcyclopentylmethoxysilane, 4-methylcyclohexyl-2-methylcyclopentylethoxysilane, 4-methylcyclohexyl-2-methylcyclopentyl-n-propoxysilane, 3,5-dimethylcyclohexyl-2-methylcyclopentylmethoxysilane, 3,5-dimethylcyclohexyl-2-methylcyclopentylethoxysilane, 3,5-dimethylcyclohexyl-2-methylcyclopentyl-n-propoxysilane, 3,3-dimethylcyclohexyl-2-methylcyclopentylmethoxysilane, 4,4-dimethylcyclohexyl-2-methylcyclopentylmethoxysilane, cyclohexyl-3-methylcyclopentylmethoxysilane, cyclohexyl-3-methylcyclopentylethoxysilane, cyclohexyl-3-methylcyclopentyl-n-propoxysilane, 3-methylcyclohexyl-3-methylcyclopentylmethoxysilane, 3-methylcyclohexyl-3-methylcyclopentylethoxysilane, 3-methylcyclohexyl-3-methylcyclopentyl-n-propoxysilane, 4-methylcyclohexyl-3-methylcyclopentylmethoxysilane, 4-methylcyclohexyl-3-methylcyclopentylethoxysilane, 4-methylcyclohexyl-3-methylcyclopentyl-n-propoxysilane, 3,5-dimethylcyclohexyl-3-methylcyclopentylmethoxysilane, 3,5-dimethylcyclohexyl-3-methylcyclopentylethoxysilane, 3,5-dimethylcyclohexyl-3-methylcyclopentyl-n-propoxysilane, 3,3-dimethylcyclohexyl-3-methylcyclopentylmethoxysilane, 4,4-dimethylcyclohexyl-3-methylcyclopentylmethoxysilane, cyclohexyl-2,3-dimethylcyclopentylmethoxysilane, cyclohexyl-2,3-dimethylcyclopentylethoxysilane, cyclohexyl-2,3-dimethylcyclopentyl-n-propoxysilane.

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Preferred of these asymmetric organosilicon compounds are cyclohexylcyclopentylidimethoxysilane, cyclohexylcyclopentyl-diethoxysilane, 3-methylcyclohexylcyclopentylidimethoxysilane, 4-methylcyclohexylcyclopentylidimethoxysilane, and 3,5-dimethylcyclohexylcyclopentylidimethoxysilane. These organosilicon compounds may be used either individually or in combination of two or more thereof.

The organosilicon compound of the present invention is useful as an (internal and/or external) electron donor for various olefin polymerization catalysts. Namely, the organosilicon compound can be used as an electron donor in the homo- or copolymerization of ethylene, propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, vinylcyclohexane, etc. In particular, the organosilicon compound is suitable for use as an electron donor of a catalyst for the homopolymerization of ethylene or propylene or the copolymerization of ethylene and propylene, and the optimal use thereof is as an electron donor of a catalyst for the homopolymerization of propylene or the copolymerization of propylene and ethylene.

The cyclohexylcyclopentylidialkoxysilane of the present invention can be prepared by various methods. In one of the simplest methods, the organosilicon compound is obtained by the reaction of a monocycloalkyltrialkoxysilane (i.e., monocyclohexyl- or monocyclopentyl-trialkoxysilane) with a cycloalkyl Grignard reagent (i.e., a Grignard reagent having a cyclopentyl or cyclohexyl group, respectively).

For example, cyclopentyl chloride (commercial product) is first reacted with magnesium in the presence of a solvent, e.g., an ether such as tetrahydrofuran, diethyl ether, or di-n-butyl ether, to yield a cyclopentyl Grignard reagent (cyclopentylmagnesium chloride). This reaction may be carried out at a temperature of from room temperature to 60°C. The cyclopentyl Grignard reagent is then reacted with cyclohexyltrimethoxysilane to obtain cyclohexylcyclopentylidimethoxysilane; this reaction may be conducted in the presence of an ether such as tetrahydrofuran, diethyl ether, or di-n-butyl ether as in the above-described

first reaction, or in the presence of an aliphatic hydrocarbon solvent such as hexane or heptane or an aromatic hydrocarbon solvent such as toluene, benzene, or xylene. This reaction may be carried out at a temperature of from 50°C to 200°C, preferably at a temperature of from 100°C to 200°C or at a temperature of from 100°C to 200°C under boiling or refluxing of the solvent.

Although the monocycloalkyltrialkoxysilane, e.g., cyclohexyltrimethoxysilane employed above, for use in the above reaction may be a commercial product, it may be prepared by various known methods. In one method, the desired compound is prepared by reacting cyclohexyltrichlorosilane with methanol to alkoxylate the silane compound with the evolution of hydrogen chloride. Although the cyclohexyltrichlorosilane for use in this reaction may be a commercial product, it may be easily prepared by the hydrosilylation reaction of cyclohexene with trichlorosilane ( $\text{HSiCl}_3$ ). Another method for preparing cyclohexyltrimethoxysilane comprises hydrogenating a commercial product of phenyltrimethoxysilane in the presence of a catalyst, e.g., a Raney nickel catalyst.

The cyclohexylcyclopentyltrimethoxysilane thus produced can be identified by nuclear magnetic resonance spectroscopy ( $^1\text{H-NMR}$ ,  $^{13}\text{C-NMR}$ ), infrared absorption spectrometry (IR), mass spectrometry (MS), etc.  $^{13}\text{C-NMR}$  spectrometry (in  $\text{CDCl}_3$ ) gives a spectrum which has a signal at  $\delta = 50.7$  attributable to the carbon atoms of the methoxy groups, signals at  $\delta = 24.5, 26.8, 26.9$ , and  $27.8$  attributable to the cyclohexyl group, and signals at  $\delta = 22.8, 26.7$ , and  $27.4$  attributable to the cyclopentyl group. IR spectrometry gives a spectrum having a peak at around  $1,100\text{ cm}^{-1}$  attributable to the Si-O-C bonds.

The organosilicon compound of the present invention, i.e., a cyclohexylcyclopentyltrialkoxysilane, when used as an electron donor serving as one component of a Ziegler-Natta catalyst for olefin polymerization, makes it possible to obtain a polyolefin having a broad molecular weight distribution and high crystallinity while retaining high performances with respect to catalytic activity and the yield of highly stereoregular polymer which performances are not lower than those conventionally known as high-performance catalysts.

Solid catalyst component (A), which constitutes the catalyst for the polymerization of olefins according to the present invention, is prepared by contacting a magnesium compound, a titanium halide compound, and an electron donor compound and contains magnesium, titanium, an internal electron donor compound and a halogen as essential components.

The magnesium compound which can be used for preparing solid catalyst component (A) includes metallic magnesium, a magnesium dihalide, a dialkylmagnesium, an alkylmagnesium halide, a dialkoxymagnesium, an alkoxymagnesium halide, a diaryloxymagnesium, and an aryloxymagnesium halide. The alkyl or alkoxy moiety of the above-described magnesium compounds generally has from 1 to 6 carbon atoms and preferably from 1 to 4 carbon atoms.

Specific examples of the magnesium halide are magnesium dichloride, magnesium dibromide, magnesium diiodide, and magnesium difluoride.

Specific examples of the dialkylmagnesium are dimethylmagnesium, diethylmagnesium, ethylmethylmagnesium, dipropylmagnesium, methylpropylmagnesium, ethylpropylmagnesium, dibutylmagnesium, butylmethylmagnesium, and butylethylmagnesium. These dialkylmagnesiums may be obtained by reacting metallic magnesium with a halogenated hydrocarbon or an alcohol.

Specific examples of the alkylmagnesium halide include ethylmagnesium chloride, propylmagnesium chloride, and butylmagnesium chloride. These alkylmagnesium halides may be obtained by reacting metallic magnesium with a halogenated hydrocarbon or an alcohol.

Specific examples of the dialkoxymagnesium and the diaryloxymagnesium include dimethoxymagnesium, diethoxymagnesium, dipropoxymagnesium, dibutoxymagnesium, diphenoxymagnesium, ethoxymethoxymagnesium, ethoxypropoxymagnesium, and butoxyethoxymagnesium.

Specific examples of the alkoxymagnesium halide are methoxymagnesium chloride, ethoxymagnesium chloride, propoxymagnesium chloride, and butoxymagnesium chloride.

Preferred of these magnesium compounds are dialkoxymagnesiums, with diethoxymagnesium and dipropoxymagnesium being especially preferred. The magnesium compounds may be used either individually or in combination of two or more thereof.

The dialkoxymagnesium, which can be used preferably, is at least one dialkoxymagnesium species having from 1 to 3 carbon atoms in the alkoxy moiety thereof and has a granular or powdered form, the particles of which may have an irregular shape or a spherical shape. In using spherical particles of diethoxymagnesium, for example, the resulting powdered polymer will have a more satisfactory particle shape and a narrower particle size distribution. As a result, the polymer powder as produced has improved handling properties, and troubles attributed to fine particles, such as obstruction, would be eliminated.

The spherical diethoxymagnesium particles as above referred to do not necessarily need to be true spheres, and ellipsoidal or potato-like particles may also be used. The terminology "spherical" as used herein may be quantified as a longer axis diameter (l) to shorter axis diameter (w) ratio (l/w) of not more

than 3, preferably from 1 to 2, and still preferably from 1 to 1.5.

The dialkoxymagnesium to be used has an average particle size of from 1 to 200  $\mu\text{m}$ , preferably from 5 to 150  $\mu\text{m}$ .

In the case of spherical diethoxymagnesium, it has an average particle size of from 1 to 100  $\mu\text{m}$ , preferably from 5 to 50  $\mu\text{m}$ , more preferably from 10 to 40  $\mu\text{m}$ . It is preferable to use particles having a sharp size distribution with a small proportion of fine or coarse particles. More specifically, particles containing not more than 20%, preferably not more than 10%, of fine particles of 5  $\mu\text{m}$  or smaller and not more than 10%, preferably not more than 5%, of coarse particles of 100  $\mu\text{m}$  or greater. Such a particle size distribution corresponds to  $\ln(D_{90}/D_{10})$  of not more than 3, preferably not more than 2, wherein  $D_{90}$  and  $D_{10}$  represent a cumulative 90% diameter and a cumulative 10% diameter, respectively, of a cumulative particle size distribution depicted from the small diameter side.

The above-mentioned dialkoxymagnesium does not always need to be present as a starting material in the preparation of solid catalyst component (A). For example, it may be prepared *in situ* from metallic magnesium and an alcohol in the presence of a catalyst, e.g., iodine at the time of preparing solid catalyst component (A).

The titanium halide compound which can be used for preparing solid catalyst component (A) is at least one of a titanium halide and an alkoxytitanium halide represented by formula:  $\text{Ti}(\text{OR}^3)_n\text{X}'_{4-n}$ , wherein  $\text{R}^3$  represents an alkyl group having from 1 to 4 carbon atoms;  $\text{X}'$  represents a chlorine atom, a bromine atom or an iodine atom; and  $n$  represents 0 or an integer of 1, 2 or 3.

Specific examples of the titanium halide include titanium tetrahalides, such as  $\text{TiCl}_4$ ,  $\text{TiBr}_4$ , and  $\text{TiI}_4$ . Specific examples of the alkoxytitanium halide are  $\text{Ti}(\text{OCH}_3)\text{Cl}_3$ ,  $\text{Ti}(\text{OC}_2\text{H}_5)\text{Cl}_3$ ,  $\text{Ti}(\text{OC}_3\text{H}_7)\text{Cl}_3$ ,  $\text{Ti}(\text{On-C}_4\text{H}_9)\text{Cl}_3$ ,  $\text{Ti}(\text{OCH}_3)_2\text{Cl}_2$ ,  $\text{Ti}(\text{OC}_2\text{H}_5)_2\text{Cl}_2$ ,  $\text{Ti}(\text{OC}_3\text{H}_7)_2\text{Cl}_2$ ,  $\text{Ti}(\text{On-C}_4\text{H}_9)_2\text{Cl}_2$ ,  $\text{Ti}(\text{OCH}_3)_3\text{Cl}$ ,  $\text{Ti}(\text{OC}_2\text{H}_5)_3\text{Cl}$ ,  $\text{Ti}(\text{OC}_3\text{H}_7)_3\text{Cl}$ , and  $\text{Ti}(\text{On-C}_4\text{H}_9)_3\text{Cl}$ . Preferred of these titanium halide compounds are titanium tetrahalides, with  $\text{TiCl}_4$  being particularly preferred. These titanium halide compounds may be used either individually or in combination of two or more thereof.

The internal electron donor compound which can be used for preparing solid catalyst component (A) is an organic compound containing oxygen or nitrogen. Such a compound include alcohols, phenols, ethers, esters, ketones, acid halides, aldehydes, amines, amides, nitriles, isocyanates, and organosilicon compounds containing an Si-O-C bond.

Specific examples of the internal electron donor compound include alcohols, such as methanol, ethanol, propanol, butanol, pentanol, hexanol, octanol, 2-ethylhexyl alcohol, and dodecanol; phenols, such as phenol and cresol; ethers, such as dimethyl ether, diethyl ether, dipropyl ether, dibutyl ether, diamyl ether, and diphenyl ether; monocarboxylic acid esters, such as methyl formate, ethyl acetate, vinyl acetate, propyl acetate, octyl acetate, cyclohexyl acetate, ethyl propionate, ethyl butyrate, methyl benzoate, ethyl benzoate, propyl benzoate, butyl benzoate, octyl benzoate, cyclohexyl benzoate, phenyl benzoate, methyl p-toluylate, ethyl p-toluylate, methyl anisate, and ethyl anisate; dicarboxylic acid esters, such as diethyl maleate, dibutyl maleate, dimethyl adipate, diethyl adipate, dipropyl adipate, dibutyl adipate, dimethyl adipate, diisodecyl adipate, dioctyl adipate, dimethyl phthalate, diethyl phthalate, dipropyl phthalate, dibutyl phthalate, dipentyl phthalate, dihexyl phthalate, diheptyl phthalate, dioctyl phthalate, dinonyl phthalate, and didecyl phthalate; ketones, such as acetone, methyl ethyl ketone, methyl butyl ketone, acetophenone, and benzophenone; acid halides, such as phthalic acid dichloride and terephthalic acid dichloride; aldehydes, such as acetaldehyde, propionaldehyde, octylaldehyde, and benzaldehyde; amines, such as methylamine, ethylamine, tributylamine, piperidine, aniline, and pyridine; amides, such as acetamide, and acrylamide; nitriles, such as acetonitrile, benzonitrile, and tolunitrile; and isocyanates, such as phenyl isocyanate, and n-butyl isocyanate.

Specific examples of the organosilicon compound containing an Si-O-C bond are trimethylmethoxysilane, trimethylethoxysilane, tri-n-propylmethoxysilane, tri-n-propylethoxysilane, tri-n-butylmethoxysilane, tri-isobutylmethoxysilane, tri-t-butylmethoxysilane, tri-n-butylethoxysilane, tricyclohexylmethoxysilane, tricyclohexylethoxysilane, dimethyldimethoxysilane, dimethyldiethoxysilane, di-n-propyldimethoxysilane, diisopropyldimethoxysilane, di-n-propyldiethoxysilane, diisopropyldiethoxysilane, di-n-butyldimethoxysilane, diisobutyldimethoxysilane, di-t-butyldimethoxysilane, di-n-butyldiethoxysilane, n-butyldimethoxysilane, bis(2-ethylhexyl)dimethoxysilane, bis(2-ethylhexyl)diethoxysilane, dicyclohexyldimethoxysilane, dicyclohexyldiethoxysilane, dicyclopentyldimethoxysilane, dicyclopentyldiethoxysilane, cyclohexylmethyldimethoxysilane, cyclohexylmethyldiethoxysilane, cyclohexylethyldimethoxysilane, cyclohexylisopropyldimethoxysilane, cyclohexylethyldiethoxysilane, cyclopentylmethyldimethoxysilane, cyclopentylethyldiethoxysilane, cyclopentylisopropyldimethoxysilane, cyclohexyl(n-pentyl)dimethoxysilane, cyclopentylisobutyldimethoxysilane, diphenyldimethoxysilane, diphenyldiethoxysilane, phenylmethyldimethoxysilane, phenylmethyldiethoxysilane, phenylethyldimethoxysilane, phenylethyldiethoxysilane, cyclohexyldimethylmethoxysilane,

cyclohexyldimethylethoxysilane, cyclohexyldiethylmethoxysilane, cyclohexyldiethylethoxysilane, 2-ethylhexyltrimethoxysilane, 2-ethylhexyltriethoxysilane, cyclohexyl(n-pentyl)diethoxysilane, cyclopentylmethylmethoxysilane, cyclopentylethylmethoxysilane, cyclopentylmethylmethoxysilane,

cyclohexyl(n-propyl)dimethoxysilane, cyclohexyl(n-butyl)dimethoxysilane, cyclohexyl(n-propyl)diethoxysilane, cyclohexyl(n-butyl)diethoxysilane, methyltrimethoxysilane, methyltriethoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, n-propyltrimethoxysilane, n-propyltriethoxysilane, isopropyltrimethoxysilane, isopropyltriethoxysilane, n-butyltrimethoxysilane, isobutyltrimethoxysilane, t-butyltrimethoxysilane, n-butyltriethoxysilane, cyclohexyltrimethoxysilane, cyclohexyltriethoxysilane, cyclopentyltrimethoxysilane, cyclopentyltriethoxysilane, vinyltrimethoxysilane, vinyltriethoxysilane, 2-ethylhexyltrimethoxysilane, 2-ethylhexyltriethoxysilane, phenyltrimethoxysilane, and phenyltriethoxysilane.

Among these internal electron donor compounds preferred are esters, with phthalic diesters being more preferred. The ester moiety in the phthalic diesters is preferably a straight chain or branched chain alkyl group having from 1 to 12 carbon atoms and preferably from 2 to 10 carbon atoms. Specific examples of suitable phthalic diesters are dimethyl phthalate, diethyl phthalate, di-n-propyl phthalate, diisopropyl phthalate, di-n-butyl phthalate, diisobutyl phthalate, ethylmethyl phthalate, methylisopropyl phthalate, ethyl-n-propyl phthalate, ethyl-n-butyl phthalate, di-n-pentyl phthalate, diisopentyl phthalate, dihexyl phthalate, di-n-heptyl phthalate, di-n-octyl phthalate, bis(2-methylhexyl)phthalate, bis(2-ethylhexyl) phthalate, di-n-nonyl phthalate, diisodecyl phthalate, bis(2,2-dimethylheptyl) phthalate, n-butylisohexyl phthalate, n-butylisooctyl phthalate, n-pentylhexyl phthalate, n-pentylisohexyl phthalate, isopentylheptyl phthalate, n-pentylisooctyl phthalate, n-pentylisononyl phthalate, isopentyl-n-decyl phthalate, n-pentylundecyl phthalate, isopentylisohexyl phthalate, n-hexylisooctyl phthalate, n-hexylisononyl phthalate, n-hexyl-n-decyl phthalate, n-heptylisooctyl phthalate, n-heptylisononyl phthalate, n-heptylneodecyl phthalate, and isooctylisononyl phthalate. These phthalic acid esters may be used either individually or in combination of two or more thereof.

Solid catalyst component (A) can be prepared by contacting the above-mentioned magnesium compound, titanium halide compound and electron donor compound in a manner appropriately selected from conventional means. Known methods for preparing a solid catalyst component are disclosed, e.g., in JP-A-63-308004, JP-A-63-314211, JP-A-64-6006, JP-A-64-14210, JP-A-64-43506, JP-A-63-3010, and JP-A-62-158704.

Typical methods for preparing solid catalyst component (A) are described below.

(1) Magnesium chloride is dissolved in a tetraalkoxytitanium, and the solution is brought into contact with polysiloxane to obtain a solid component. The solid component is reacted with silicon tetrachloride, contacted with phthalic acid chloride, and reacted with titanium tetrachloride to prepare solid catalyst component (A). The resulting solid catalyst component may be preliminarily treated with an organoaluminum compound, an organosilicon compound, and an olefin.

(2) Anhydrous magnesium chloride and 2-ethylhexyl alcohol are reacted to form a uniform solution, which is brought into contact with phthalic anhydride. The resulting solution is then brought into contact with titanium tetrachloride and diester of phthalic acid to obtain a solid component, which is further reacted with titanium tetrachloride to prepare solid catalyst component (A).

(3) Metallic magnesium, butyl chloride, and butyl ether are reacted to synthesize an organomagnesium compound. The organomagnesium compound is brought into contact with tetrabutoxytitanium and tetraethoxysilane to obtain a solid product, which is then brought into contact with a diester of phthalic acid, dibutyl ether, and titanium tetrachloride to prepare solid catalyst component (A). The resulting solid catalyst component may be preliminarily treated with an organoaluminum compound, an organosilicon compound, and an olefin.

(4) An organomagnesium compound, e.g., dibutylmagnesium, and an organoaluminum compound are brought into contact with an alcohol, e.g., butanol or 2-ethylhexyl alcohol, in the presence of a hydrocarbon solvent to form a uniform solution. The resulting solution is brought into contact with a silicon compound, e.g.,  $\text{SiCl}_4$ ,  $\text{HSiCl}_3$  or polysiloxane, to obtain a solid component. The solid component is brought into contact with titanium tetrachloride and a diester of phthalic acid in the presence of an aromatic hydrocarbon solvent, and the reaction mixture is further brought into contact with titanium tetrachloride to obtain solid catalyst component (A).

(5) Magnesium chloride, a tetraalkoxytitanium, and an aliphatic alcohol are brought into contact in the presence of an aliphatic hydrocarbon to form a uniform solution. Titanium tetrachloride is added to the solution, and the mixture is heated to precipitate a solid component. The solid component is contacted with a diester of phthalic acid and further reacted with titanium tetrachloride to prepare solid catalyst component (A).

(6) Metallic magnesium powder, an alkyl monohalide, and iodine are contacted. The resulting reaction product, a tetraalkoxytitanium, an acid halide, and an aliphatic alcohol are contacted in the presence of an aliphatic hydrocarbon to form a uniform solution. Titanium tetrachloride is added to the solution, and the mixture is heated to precipitate a solid component. The solid component is brought into contact with a diester of phthalic acid and further reacted with titanium tetrachloride to prepare solid catalyst component (A).

(7) Diethoxymagnesium is suspended in an alkylbenzene or a halogenated hydrocarbon solvent, and the resulting suspension is brought into contact with titanium tetrachloride. The mixture is heated and then contacted with a diester of phthalic acid to obtain a solid component. The solid component is washed with an alkylbenzene and again contacted with titanium tetrachloride in the presence of the alkylbenzene to prepare solid catalyst component (A). The resulting solid catalyst component may be subjected to a heat treatment in the presence or absence of a hydrocarbon solvent.

(8) Diethoxymagnesium is suspended in an alkylbenzene, and the resulting suspension is brought into contact with titanium tetrachloride and phthalic acid chloride to obtain a solid component. The solid component is washed with an alkylbenzene and again contacted with titanium tetrachloride in the presence of the alkylbenzene to prepare solid catalyst component (A). The resulting solid catalyst component may further be contacted with titanium tetrachloride twice or more times.

(9) Diethoxymagnesium, calcium chloride, and a silicon compound represented by  $\text{Si}(\text{OR}^4)_4$  (wherein  $\text{R}^4$  is an alkyl group or an aryl group) are co-ground, and the resulting grinds are suspended in an aromatic hydrocarbon. The suspension is brought into contact with titanium tetrachloride and an diester of aromatic dicarboxylic acid, and the product is further contacted with titanium tetrachloride to prepare solid catalyst component (A).

(10) Diethoxymagnesium and a diester of phthalic acid are suspended in an alkylbenzene, and the suspension is added to titanium tetrachloride to obtain a solid component. The solid component is washed with an alkylbenzene, and further contacted with titanium tetrachloride in the presence of the alkylbenzene to prepare solid catalyst component (A).

(11) A calcium halide and aliphatic magnesium, e.g., magnesium stearate, are contact reacted with titanium tetrachloride and an aromatic dicarboxylic acid diester, and the reaction product is further brought into contact with titanium tetrachloride to prepare solid catalyst component (A).

(12) Diethoxymagnesium is suspended in an alkylbenzene or a halogenated hydrocarbon solvent, and the resulting suspension is brought into contact with titanium tetrachloride, and the mixture is heated and contacted with a diester of phthalic acid to react. The resulting solid component is washed with an alkylbenzene and further contacted with titanium tetrachloride in the presence of the alkylbenzene to prepare solid catalyst component (A). At any stage of the above preparation procedure, the system may be brought into contact with aluminum chloride.

(13) Diethoxymagnesium is suspended in an alkylbenzene or a halogenated hydrocarbon solvent, and the resulting suspension is brought into contact with titanium tetrachloride, and the mixture is heated and contacted with two or more diesters of phthalic acid different in the carbon atom number of the alkyl moiety to obtain a solid component. The resulting solid component is washed with an alkylbenzene and further contacted with titanium tetrachloride in the presence of the alkylbenzene to prepare solid catalyst component (A). In the above preparation, when the solid component is brought into contact with titanium tetrachloride, it may again contacted with two or more diesters of phthalic acid different in the carbon atom number of the alkyl moiety. Further, the diesters of phthalic acid may be used in combination with the above-enumerated electron donor compound other than diesters of phthalic acid.

(14) Diethoxymagnesium, titanium tetrachloride, and a diester of phthalic acid are brought into contact in the presence of chlorobenzene, and the reaction product is then contacted with titanium tetrachloride and phthalic acid dichloride. The product is further contact reacted with titanium tetrachloride to prepare solid catalyst component (A). The thus prepared solid catalyst component may further be contacted with titanium tetrachloride. Further, at any stage of the above preparation procedure, a silicon compound may be contacted with the preparation system.

(15) Diethoxymagnesium, 2-ethylhexyl alcohol, and carbon dioxide are brought into contact in the presence of toluene to form a uniform solution. The solution is contacted with titanium tetrachloride and a diester of phthalic acid to obtain a solid component. The solid component is dissolved in tetrahydrofuran, and the solid component is made to precipitate. The resulting solid component is contact reacted with titanium tetrachloride to prepare solid catalyst component. If desired, the contact with titanium tetrachloride may be conducted repeatedly. At any stage of the above preparation procedure, a silicon compound, e.g., tetrabutoxysilane, may be contacted with the preparation system.

The amounts of the magnesium compound, titanium halide compound and electron donor compound to be used for the preparation of solid catalyst component (A) vary depending on the method of preparation and cannot be generally specified. For example, the titanium halide compound is used in an amount of from 0.5 to 100 mol, preferably from 1 to 10 mol, and the electron donor compound from 0.01 to 3 mol, preferably from 0.02 to 1 mol, each per mole of the magnesium compound. The titanium content in solid catalyst component (A) is not particularly limited and it is generally from 0.5 to 10 % by weight, preferably from 1 to 5 % by weight, based on the weight of solid catalyst component (A).

Organoaluminum compound (B) which can be used in the present invention includes compounds represented by  $R^5_yAlY_{3-y}$ , wherein  $R^5$  represents a straight chain or branched chain alkyl group, preferably having from up to 15 carbon atoms, more preferably up to 5 carbon atoms; Y represents a hydrogen atom, a halogen atom, or an alkoxyl group preferably having up to 4 carbon atoms; and y represents a real number of 1 to 3.

Specific examples of organoaluminum compound (B) include trialkylaluminums such as triethylaluminum and triisobutylaluminum; dialkylaluminum halides such as diethylaluminum bromide, diethylaluminum chloride and dibutylaluminum chloride; alkylaluminum sesquihalides such as ethylaluminum sesquichloride and butylaluminum sesquichloride; alkylaluminum alkoxides such as diethylaluminum ethoxide and dibutylaluminum butoxide; alkylaluminum hydrides such as diethylaluminum hydride, dibutylaluminum hydride and ethylaluminum hydride; and a mixture thereof. Of these, triethylaluminum and triisobutylaluminum are preferred.

In the present invention, an olefin is homo- or copolymerized in the presence of a catalyst comprising solid catalyst component (A), organoaluminum compound (B), and organosilicon compound (C). The ratio of components (A), (B), and (C) to be used is not particularly limited as long as the effects of the present invention are not impaired. Usually, organoaluminum compound (B) is used in an amount of from 1 to 500 mol and preferably from 5 to 400 mol per mol of the titanium atom in solid catalyst component (A), and organosilicon compound (C) is used in an amount of from 0.0020 to 2 mol, preferably from 0.0020 to 0.5 mol, and more preferably from 0.0025 to 0.5 mol per mol of organoaluminum compound (B).

The catalyst of the present invention can be prepared by bringing the above-described components (A), (B) and (C) into contact. There is no particular limitation on the order in contact of the components (A), (B) and (C). In general, the component (B) is brought into contact with the component (C) and subsequently with the component (A), or the component (B) is brought into contact with the component (A) and subsequently with the component (C).

Recommended combinations of the components (A), (B), and (C) are tabulated in Table 1 below.

TABLE 1

Solid Catalyst Component (A) (Process of Preparation)	Organoaluminum Compound (B)	Organosilicon Compound (C)
process (7)	triethylaluminum	cyclohexylcyclopentylmethoxysilane
process (7)	triethylaluminum	3-methylcyclohexylcyclopentylmethoxysilane
process (8)	triethylaluminum	cyclohexylcyclopentylmethoxysilane
process (10)	triethylaluminum	cyclohexylcyclopentylmethoxysilane
process (10)	triethylaluminum	4-methylcyclohexylcyclopentylmethoxysilane
process (12)	triethylaluminum	cyclohexylcyclopentylmethoxysilane
process (12)	triethylaluminum	3-methylcyclohexylcyclopentylmethoxysilane
process (13)	triethylaluminum	cyclohexylcyclopentylmethoxysilane
Process (13)	triethylaluminum	3,5-dimethylcyclohexylcyclopentylmethoxysilane

Of the above-mentioned solid catalyst components (A), those described below as embodiments 1 and 2 are preferred as a component of the catalyst for polymerization of olefins since they can provide polyolefins particularly having a broad molecular weight distribution and a high stereoregularity at a high yield. Besides, a solid catalyst component of embodiment 1 as described below can provide a stereoregular polymer having a density of from 0.900 to 0.906 g/ml at a low RDS and yet at a high yield, with suppressing formation of fine powders.

The solid catalyst component of embodiment 1 is prepared using substances (a) to (d) and optionally substance (e):

(a) a dialkoxymagnesium represented by  $Mg(OR^6)_2$  wherein  $R^6$  represents a straight chain or branched chain alkyl group or an aryl group;

(b) an aluminium compound represented by  $\text{Al}(\text{OR}^7)_m\text{X}^2_{3-m}$  wherein  $\text{R}^7$  represents a straight chain or branched chain alkyl group,  $\text{X}^2$  represents a halogen atom, and  $m$  is 0 or an integer of 1 to 3;

(c) titanium tetrachloride;

(d) a diester of aromatic dicarboxylic acid; and

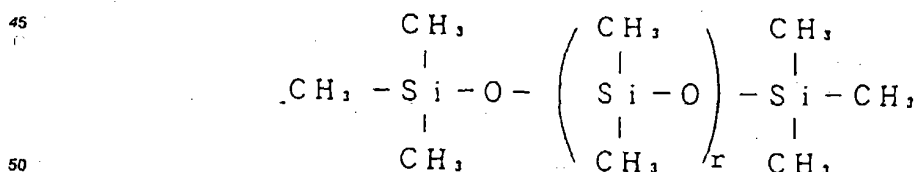
5 (e) a dimethyl polysiloxane.

The dialkoxymagnesium (substance (a)) represented by  $\text{Mg}(\text{OR}^6)_2$  used in the preparation of the solid catalyst component of embodiment 1 preferably has a straight chain or branched chain alkyl group having up to 10 carbon atoms, more preferably 2 to 4 carbon atoms, or an aryl group for  $\text{R}^6$ . Examples of substance (a) includes diethoxymagnesium, dipropoxymagnesium, dibutoxymagnesium, and diphenoxymagnesium, with diethoxymagnesium being preferred. Details of the dialkoxymagnesium, which can be used preferably, are the same as described above.

The aluminium compound represented by  $\text{Al}(\text{OR}^7)_m\text{X}^2_{3-m}$  (substance (b)) used in the preparation of the solid catalyst component of embodiment 1 preferably has a straight chain or branched chain alkyl group having up to 10 carbon atoms, more preferably up to 5 carbon atoms, for  $\text{R}^7$ , such as trihalogenated aluminium, halogenated alkoxyaluminium and trialkoxyaluminium. Examples of trihalogenated aluminium include aluminium trichloride, aluminium tribromide and aluminium triiodide, with aluminium trichloride being preferred. Examples of halogenated alkoxyaluminium includes diethoxychloroaluminium, ethoxydichloroaluminium, diisopropoxychloroaluminium, isopropoxydichloroaluminium, dibutoxychloroaluminium, and butoxydichloroaluminium, with ethoxydichloroaluminium, diisopropoxychloroaluminium, and isopropoxydichloroaluminium being preferred. Examples of trialkoxyaluminium include trimethoxyaluminium, triethoxyaluminium, tripropoxyaluminium, triisopropoxyaluminium, tributoxyaluminium, and triisobutoxyaluminium, with triethoxyaluminium and triisopropoxyaluminium being preferred. These aluminium compounds may be used independently or as admixture thereof.

The aromatic dicarboxylic diester substance (d) used in the preparation of the solid catalyst component of embodiment 1 is preferably a phthalic diester. The ester moiety thereof is preferably a straight chain or branched chain alkyl group having up to 15 carbon atoms, more preferably 2 to 12 carbon atoms. Examples of phthalic diester include dimethyl phthalate, diethyl phthalate, di-n-propyl phthalate, di-isopropyl phthalate, di-n-butyl phthalate, di-iso-butyl phthalate, ethylmethyl phthalate, butylethyl phthalate, methyl(iso-propyl) phthalate, ethyl-n-propyl phthalate, ethyl-n-butyl phthalate, di-n-pentyl phthalate, di-iso-pentyl phthalate, di-n-hexyl phthalate, di-n-heptyl phthalate, di-n-octyl phthalate, bis(2-methylhexyl) phthalate, bis(2-ethylhexyl) phthalate, di-n-nonyl phthalate, di-iso-decyl phthalate, bis(2,2-dimethylheptyl) phthalate, n-butyl(iso-hexyl) phthalate, ethyl(iso-octyl) phthalate, n-butyl(iso-octyl) phthalate, n-pentyl(n-hexyl) phthalate, n-pentyl(iso-hexyl) phthalate, iso-pentyl(n-heptyl) phthalate, n-pentyl(iso-octyl) phthalate, n-pentyl(iso-nonyl) phthalate, iso-pentyl(n-decyl) phthalate, n-pentyl(n-undecyl) phthalate, iso-pentyl(iso-hexyl) phthalate, n-hexyl(iso-octyl) phthalate, n-hexyl(iso-nonyl) phthalate, n-hexyl(n-decyl) phthalate, n-heptyl(iso-octyl) phthalate, n-heptyl(isononyl) phthalate, n-heptyl(neo-decyl) phthalate, and iso-octyl(isononyl) phthalate. At least one of them is used. Of these, diethyl phthalate, di-n-propyl phthalate, di-n-butyl phthalate, di-iso-butyl phthalate, and bis(2-ethylhexyl) phthalate are preferred. These phthalic diesters may be used independently or as admixture thereof.

40 The dimethyl polysiloxane (e) (substance (e)) is an optional component constituting the solid catalyst component of embodiment 1 and is preferably a polymer silicone compound represented by the following formula



wherein  $r$  is 1,000 to 30,000. The dimethyl polysiloxane has a viscosity of 5 to 1,000 centistokes, preferably 10 to 500 centistokes, at a room temperature.

55 The solid catalyst component of embodiment 1 can be prepared by contacting the substance (a), the substance (b), titanium tetrachloride referred to as the "substance (c)" and the substance (d); or the substance (a), the substance (b), the substance (c), the substance (d) and the substance (e), in the presence or absence of an inert organic solvent. For easy operation, the processing is preferably carried

out in the presence of the solvent. The inert organic solvent used includes saturated hydrocarbon such as hexane, heptane and cyclohexane; aromatic hydrocarbon such as benzene, toluene, xylene and ethylbenzene; and halogenated hydrocarbon such as o-dichlorobenzene, methylene chloride, carbon tetrachloride, and dichloroethane. Among these, aromatic hydrocarbons having a boiling point of 90 to 150 °C are preferably used.

The used amount of the respective substances are 0.01 to 10 g, preferably 0.05 to 2.0 g for the substance (b), 0.1 to 200 ml, preferably 0.5 to 100 ml for the substance (c), 0.01 to 1.0 g, preferably 0.1 to 0.5 g for the substance (d), and 0.05 to 5.0 ml, preferably 0.1 to 1.0 ml for the substance (e), each based on 1 g of the substance (a). The amount of the inert organic solvent is not specifically limited, and it is preferably from 1 to 10 ml per 1 ml of the substance (c) for easy operation.

Each of these substances (a) to (e) can be brought into contact at once or piece by piece. The contact of the respective substances is carried out in a vessel equipped with a stirrer under an inert gas atmosphere in a moisture-free condition while stirring. The contact temperature may be a relatively low temperature close to room temperature in the case of simply contacting by way of stirring and mixing the substances or in case of dispersing or suspending the substances in an inert organic solvent. In case of reacting the substances after contact to obtain a reaction product, the temperature is preferably from 40 to 130 °C. If the temperature is less than 40 °C, the reaction does not sufficiently proceed, and the resulting solid catalyst component may not exhibit sufficient catalytic performances. If the temperature is higher than 130 °C, the solvent is markedly evaporated so that the reaction becomes unstable. The reaction time is preferably 1 minute or more, more preferably 10 minutes or more, and most preferably 30 minutes or more.

Preferred contact orders of the respective substances in the present invention are as follows:

- (1) the substances (a), (b), (c) and (d) are contacted at the same time;
- (2) the substance (c) is contacted to a product obtained by contacting the substances (a), (b), (c) and (d);
- (3) the substance (d) is contacted to a product obtained by contacting the substances (a), (b) and (c) in advance;
- (4) the substance (d) is contacted to a product obtained by contacting the substances (a), (b) and (c) in advance, and then the substance (c) is further contacted;
- (5) the substance (b) is contacted to a product obtained by contacting the substances (a), (c) and (d) in advance;
- (6) the substance (b) is contacted to a product obtained by contacting the substances (a), (c) and (d) in advance, and then the substance (c) is further contacted;
- (7) the substance (b) is contacted to a product obtained by contacting the substances (a), (c) and (d) in advance, and then the substances (b) and (c) are further contacted;
- (8) the substances (b) and (c) are contacted to a product obtained by contacting the substances (a), (b), (c) and (d) in advance;
- (9) the substances (a), (b), (c), (d) and (e) are contacted at the same time;
- (10) the substance (c) is contacted to a product obtained by contacting the substances (a), (b), (c), (d) and (e);
- (11) the substances (d) and (e) are contacted to a product obtained by contacting the substances (a), (b) and (c) in advance;
- (12) the substances (d) and (e) are contacted to a product obtained by contacting the substances (a), (b) and (c) in advance, and then the substance (c) is further contacted;
- (13) the substance (b) is contacted to a product obtained by contacting the substances (a), (c), (d) and (e) in advance;
- (14) the substance (b) is contacted to a product obtained by contacting the substances (a), (c), (d) and (e) in advance, and then the substance (c) is further contacted;
- (15) the substance (b) is contacted to a product obtained by contacting the substances (a), (c), (d) and (e) in advance, and then the substances (b) and (c) are further contacted; and
- (16) the substances (b) and (c) are contacted to a product obtained by contacting the substances (a), (b), (c), (d) and (e).

The contact condition in the case of contacting the substance (b) and/or the substance (c) in the later stage is preferably that the substance (b) and/or the substance (c) are added as they are, or diluted by the inert organic solvent described above and then added (the latter being preferably) to the product, and allowed to stand at a temperature of 40 to 130 °C for 1 minute or more, preferably 10 minutes or more, and more preferably 30 minutes or more. A ratio of the substance (b) to the substance (c), when both are added to the product, may be the same or different as that in the contact and reaction at the former stage (to form the product). The product obtained by the contact and reaction in the former stage may be washed with the inert organic solvent described above, and again contacted with the substance (b) and/or the substance (c).

By the use of the solid catalyst component of embodiment 1 of the present invention, in polymerization of olefins, particularly polymerization of propylene, a stereoregular polymer having a low density of 0.900 to 0.906 g/ml can be produced stably at a low RDS of 3% or less (preferably 2% or less). Further, the yield of the polymer per unit amount of the resulting catalyst is high, that is, the resulting catalyst has a high polymerization activity and exhibits excellent performances in terms of catalytic life and reduction of a fine powder content in the resulting polymer.

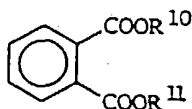
Next, the solid catalyst component of embodiment 2 will be explained below.

The solid catalyst component of embodiment 2 is prepared by contacting compounds (a') to (d'):

(a') a magnesium compound represented by  $Mg(OR^8)_2 \cdot pX^3$  (wherein each  $R^8$  independently represents an alkyl group having 1 to 4 carbon atoms or a phenyl group,  $X^3$  represents chlorine or bromine, and  $p$  represents 0 or 1);

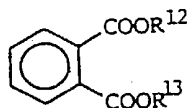
(b') a titanium compound represented by  $Ti(OR^9)_q X^4_{4-q}$  (wherein  $R^9$  represents an alkyl group having 1 to 4 carbon atoms,  $X^4$  represents a halogen element, and  $q$  is 0, 1, 2 or 3)

(c') at least one phthalic diester represented by



(wherein  $R^{10}$  and  $R^{11}$  may be the same or different and each represents an alkyl group having 1 to 4 carbon atoms); and

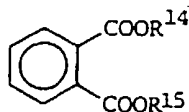
(d') at least one phthalic diester represented by



(wherein  $R^{12}$  and  $R^{13}$  may be the same or different and each represents an alkyl group having 4 to 12 carbon atoms, provided that the total number of carbon atoms of  $R^{12}$  and  $R^{13}$  is from 10 to 20).

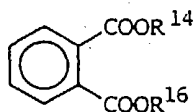
The solid catalyst component of embodiment 2 for olefin polymerization contains the following phthalic diester compounds (e') to (g'):

(e')



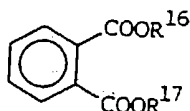
(wherein  $R^{14}$  and  $R^{15}$  may be the same or different and each represents an alkyl group having 1 to 4 carbon atoms);

(f')



(wherein  $R^{16}$  represents an alkyl group having 5 to 12 carbon atoms);

(g')



(wherein R<sup>17</sup>

represents an alkyl group having 4 to 12 carbon atoms, provided that the total number of carbon atoms of R<sup>16</sup> and R<sup>17</sup> is from 10 to 20).

As described above, the solid catalyst component of embodiment 2 is prepared by contacting the magnesium compound of compound (a'), the titanium compound of compound (b'), the phthalic diester of compound (c'), and the phthalic diester of compound (d'), and contains the phthalic diester of compound (e'), the phthalic diester of compound (f'), and the phthalic diester of compound (g').

Examples of the magnesium compound of compound (a') include dialkoxymagnesiums such as dimethoxymagnesium, diethoxymagnesium, di-n-propoxymagnesium, diisopropoxymagnesium, di-n-butoxymagnesium, diisobutylmagnesium, methoxyethoxymagnesium, methoxypropoxymagnesium, methoxy-n-butoxymagnesium, and ethoxy-n-propoxymagnesium; diaryloxymagnesiums such as diphenoxymagnesium; alkoxymagnesium halides such as methoxymagnesium chloride, ethoxymagnesium chloride, methoxymagnesium bromide, and ethoxymagnesium bromide; and aryloxymagnesium halides such as phenoxymagnesium chloride and phenoxymagnesium bromide. Of these, dimethoxymagnesium, diethoxymagnesium, and dipropoxymagnesium, are preferred. In particular, diethoxymagnesium is preferably used. These magnesium compounds may be used alone or in combination of two or more thereof as compound (a').

The magnesium compound (a') is preferably a particulate compound which has an average particle diameter of from 10 to 150  $\mu\text{m}$  and contains no fine particles of 1  $\mu\text{m}$  or smaller. Such particle properties are preferred in that the solid catalyst component obtained using this magnesium compound has a reduced fine-particle content, and that the use of this catalyst component not only yields a polymer having a reduced content of fine polymer particles but also leads to an improvement in catalytic activity.

Compound (b') is at least one titanium compound selected from titanium tetrahalides and alkoxytitanium halides. Examples of the titanium tetrahalides include  $\text{TiCl}_4$ ,  $\text{TiBr}_4$ , and  $\text{TiI}_4$ , and examples of the alkoxytitanium halides include  $\text{Ti}(\text{OCH}_3)\text{Cl}_3$ ,  $\text{Ti}(\text{OC}_2\text{H}_5)\text{Cl}_3$ ,  $\text{Ti}(\text{OC}_3\text{H}_7)\text{Cl}_3$ ,  $\text{Ti}(\text{O-n-C}_4\text{H}_9)\text{Cl}_3$ ,  $\text{Ti}(\text{OCH}_3)_2\text{Cl}_2$ ,  $\text{Ti}(\text{OC}_2\text{H}_5)_2\text{Cl}_2$ ,  $\text{Ti}(\text{OC}_3\text{H}_7)_2\text{Cl}_2$ ,  $\text{Ti}(\text{O-n-C}_4\text{H}_9)_2\text{Cl}_2$ ,  $\text{Ti}(\text{OCH}_3)_3\text{Cl}$ ,  $\text{Ti}(\text{OC}_2\text{H}_5)_3\text{Cl}$ ,  $\text{Ti}(\text{OC}_3\text{H}_7)_3\text{Cl}$ , and  $\text{Ti}(\text{O-n-C}_4\text{H}_9)_3\text{Cl}$ . These titanium compounds are used alone or in combination of two or more thereof. Preferred of these are the titanium tetrahalides, especially  $\text{TiCl}_4$ .

At least one phthalic diester of compound (c') (hereinafter often referred to as "first ester") and at least one phthalic diester of compound (d') (hereinafter referred to as "second ester") are used. Namely, at least two kinds of phthalic diesters are used.

Examples of the first ester include dimethyl phthalate, diethyl phthalate, di-n-propyl phthalate, diisopropyl phthalate, di-n-butyl phthalate, diisobutyl phthalate, ethyl methyl phthalate, methyl isopropyl phthalate, ethyl n-propyl phthalate, and ethyl n-butyl phthalate. These esters may be used alone or in combination of two or more thereof.

Examples of the second ester include di-n-pentyl phthalate, diisopentyl phthalate, dihexyl phthalate, di-n-heptyl phthalate, di-n-octyl phthalate, bis(2-methylhexyl) phthalate, bis(2-ethylhexyl) phthalate, di-n-nonyl phthalate, diisodecyl phthalate, bis(2,2-dimethylheptyl) phthalate, n-butyl isohexyl phthalate, n-butyl isooctyl phthalate, n-pentyl hexyl phthalate, n-pentyl isohexyl phthalate, isopentyl heptyl phthalate, n-pentyl isooctyl phthalate, n-pentyl isononyl phthalate, isopentyl n-decyl phthalate, n-pentyl undecyl phthalate, isopentyl isohexyl phthalate, n-hexyl isooctyl phthalate, n-hexyl isononyl phthalate, n-hexyl n-decyl phthalate, n-heptyl isooctyl phthalate, n-heptyl isononyl phthalate, n-heptyl neodecyl phthalate, and isooctyl isononyl phthalate. These esters may be used alone or in combination of two or more thereof.

Combinations of the first ester and the second ester are not particularly limited. However, preferred combinations are those in which the difference between the total number of carbon atoms of R<sup>16</sup> and R<sup>17</sup> in the first ester and the total number of carbon atoms of R<sup>12</sup> and R<sup>13</sup> in the second ester is 6 or larger, preferably from 8 to 18, preferably from 10 to 18, especially preferably from 10 to 16. Examples of such preferred combinations are given in Table 2.

TABLE 2

	First Ester	Second Ester
5	(1) Diethyl phthalate	Di-n-pentyl phthalate
	(2) Diethyl phthalate	Diphthalate of C <sub>6-12</sub> n-alcohol
	(3) Diethyl phthalate	Bis(2-ethylhexyl) phthalate
	(4) Dimethyl phthalate	Diisopentyl phthalate
	(5) Dimethyl phthalate	Bis(2-ethylhexyl) phthalate
10	(6) Di-n-propyl phthalate	Bis(2-methylhexyl) phthalate + bis(2-ethylhexyl) phthalate
	(7) Diethyl phthalate + di-n-butyl phthalate	Bis(2-ethylhexyl) phthalate
	(8) Dimethyl phthalate + diethyl phthalate	Bis(2-ethylhexyl) phthalate
	(9) Dimethyl phthalate + diethyl phthalate	Bis(2-ethylhexyl) phthalate + di-n-hexyl phthalate
15	(10) Diethyl phthalate + diisobutyl phthalate	Bis(2-ethylhexyl) phthalate + di-n-hexyl phthalate

In preparing the solid catalyst component of embodiment 2, it is necessary to use a combination of at least one of the above-enumerated first esters and at least one of the above-enumerated second esters. These first and second esters may be used in combination with one or more other electron-donor compounds. Examples of such electron-donor compounds include other phthalic diester compounds, mono- or diesters of cyclohexanecarboxylic acids, esters of aromatic monocarboxylic acids, (e.g., benzoic acid), diesters of dicarboxylic acids, (e.g., adipic acid), carbonic esters, and ethers. Examples of preferred combinations are given in Table 3.

TABLE 3

	First Ester	Second Ester	Other Electron-donating compound
25	(1) Diethyl phthalate	Di-n-octyl phthalate	Ethyl benzoate
	(2) Diethyl phthalate	Bis(2-ethylhexyl) phthalate	Ethyl p-toluate
30	(3) Di-n-propyl phthalate	Bis(2-ethylhexyl) phthalate	Diethyl carbonate
	(4) Diethyl phthalate	Bis(2-ethylhexyl) phthalate	Diisodecyl adipate
	(5) Diethyl phthalate	Bis(2-ethylhexyl) phthalate	Diethyl ether

The solid catalyst component of embodiment 2 is obtained by contacting a combination of at least one first ester and at least one second ester, which esters differ from each other in carbon number, with the Mg compound and the Ti compound. This solid catalyst component functions to enable a polyolefin to have not only a significantly broadened molecular weight distribution but greatly improved crystallinity.

In preparing the solid catalyst component of this embodiment by contacting the above-described compounds (a') to (d'), these compounds may be contacted in the absence of an inert organic solvent. However, from the standpoints of reaction control and easiness of operation, it is preferred to contact the compounds in the presence of an inert organic solvent. Examples of the inert organic solvent include saturated hydrocarbons, e.g., hexane, heptane, and cyclohexane; aromatic hydrocarbons, e.g., benzene, toluene, xylene, and ethylbenzene; and halogenated hydrocarbons, e.g., o-dichlorobenzene, methylene chloride, carbon tetrachloride, and dichloroethane. Of these, benzene, toluene, xylene, and ethylbenzene are preferred, which are aromatic hydrocarbons having a boiling point of about from 90°C to 150°C. The amount of such an inert organic solvent is not particularly limited, but it is preferably such that the proportion of the solvent to the titanium compound of compound (b') is from 0.1 to 10 by volume.

The sequence of the contacting of the compounds used is not limited. Examples of the sequence of contacting are shown in Table 4. It is, however, preferred that the first ester of compound (c') be contacted at a lower temperature than the second ester of compound (d'). Consequently, compound (a') or a mixture of compounds (a') and (b') should be first contacted with compound (c'), and is then contacted with compound (d') desirably at a higher temperature. Each compound may be contacted repeatedly. In particular, repeatedly contacting compound (b') or (c') enhances the function of producing a polymer having a broadened molecular weight distribution and heightened crystallinity.

TABLE 4

(1)	$(a') + (b') \rightarrow (c') \rightarrow (d')$
(2)	$(a') + (b') \rightarrow (c') \rightarrow (d') \rightarrow (b')$
(3)	$(a') + (b') \rightarrow (c') \rightarrow (d') \rightarrow (b') \rightarrow (b')$
(4)	$(a') + (b') \rightarrow (c') \rightarrow (d') \rightarrow (c') \rightarrow (b')$
(5)	$(a') + (b') \rightarrow (c') \rightarrow (d') \rightarrow (b') \rightarrow (c')$
(6)	$(a') + (b') \rightarrow (c') \rightarrow (d') \rightarrow (b') \rightarrow (d')$
(7)	$(a') + (b') \rightarrow (c') \rightarrow (d') \rightarrow (c') \rightarrow (d') \rightarrow (b')$
(8)	$(a') + (b') \rightarrow (c') \rightarrow (d') \rightarrow (b') \rightarrow (c') \rightarrow (d') \rightarrow (b')$
(9)	$(a') + (c') \rightarrow (b') \rightarrow (d')$
(10)	$(a') + (c') \rightarrow (b') \rightarrow (d') \rightarrow (b')$
(11)	$(a') + (c') \rightarrow (b') \rightarrow (b') \rightarrow (d')$
(12)	$(a') + (c') \rightarrow (d') \rightarrow (b') \rightarrow (b')$
(13)	$(a') + (c') \rightarrow (d') \rightarrow (b') \rightarrow (b') \rightarrow (c')$

The following are examples of methods for preparing the solid catalyst component of embodiment 2.

Method 1: Compound (a') and compound (b') are contacted with each other at a temperature of from -20 to 100 °C, preferably from -10 to 70 °C, more preferably from 0 to 30 °C, in an inert organic solvent such as toluene, and then reacted at a temperature of from 0 to 130 °C, preferably from 70 to 120 °C. Compound (c') is then added thereto at a temperature of from -20 to 130 °C, preferably from 30 to 70 °C, to contact the ingredients together. Thereafter, compound (d') is added thereto at a temperature of from -20 to 130 °C, preferably from 10 to 130 °C, more preferably from 80 to 115 °C, to contact the ingredients together. In this method, compound (c') is preferably contacted at a lower temperature than compound (d'). Compound (c') and/or compound (d') may be repeatedly added as shown in Table 4, (4) to (8). It is also possible to change the temperature at which compound (c') or (d') is added and reacted. For example, the first addition of compound (c') is conducted at a temperature of from -10 to 10 °C and the second addition thereof is conducted at a temperature of from 40 to 70 °C, while the first addition of compound (d') is conducted at a temperature of from 40 to 70 °C and the second addition thereof is conducted at a temperature of from 80 to 150 °C.

Method 2: As shown in Table 3, (9) to (11), compound (a') is first contacted with compound (c'). This contacting is performed in an inert organic solvent at a temperature of from -20 to 70 °C, preferably from 0 to 50 °C, more preferably from 0 to 30 °C. Compound (b') is then contacted at a temperature of from -20 to 100 °C, preferably from 0 to 30 °C, more preferably from 10 to 30 °C. Thereafter, compound (d') is contacted at a temperature of from -20 to 130 °C, preferably from 10 to 130 °C, more preferably from 80 to 115 °C, and the ingredients are then reacted at a temperature of from 0 to 130 °C, preferably from 70 to 120 °C. In this method also, compound (c') and (d') may be added repeatedly. Further, the temperature at which compound (c') or (d') is added and reacted may be changed. For example, compound (c') is first added to the organic solvent containing compound (a') at a temperature of from -10 to 10 °C and the second addition of compound (c') is conducted at a higher temperature, e.g., from 40 to 70 °C. On the other hand, compound (d') is first added at a temperature of from 40 to 70 °C and the second addition thereof is conducted at a temperature of from 80 to 115 °C.

Method 3: As shown in Table 3, (12) and (13), compound (a') is first contacted with compound (c') and compound (d'). This contacting is performed in an organic solvent at a temperature of from -20 to 120 °C, preferably from 0 to 100 °C, more preferably from 10 to 70 °C. Compound (b') is then contacted at a temperature of from -20 to 100 °C, preferably from 0 to 30 °C, more preferably from 10 to 60 °C. After the contacting of the compounds, the mixture is heated to a temperature of from 0 to 130 °C, preferably from 70 to 120 °C, to react the compounds. Thereafter, compound (c') and/or compound (d') is further added and reacted at a temperature different from the temperature for the first addition thereof. For example, the first addition of compound (c') and/or compound (d') is performed at a temperature of from 10 to 30 °C and the second addition thereof is performed at a temperature of from 30 to 70 °C.

The duration for which compounds are contacted in each contacting operation is not particularly limited, but it is generally from 30 minutes to 5 hours, desirably from 1 to 3 hours. The intermediate reaction product formed by each contacting of compounds may be isolated and washed with an inert organic solvent, before being subjected to subsequent contacting with another compound. Further, the repeated contacting of compound (b') is preferred in improving the performances of the catalyst component, and is effective especially in widening the molecular weight distribution of the polymer to be yielded and

increasing the crystallinity thereof.

The proportions of the compounds to be used cannot be specified unconditionally because they vary depending on preparation method. In general, however, the amount of compound (b') may be from 0.5 to 100 mol, preferably from 1 to 10 mol, that of compound (c') may be from 0.01 to 0.5 mol, preferably from 0.02 to 0.2 mol, and that of compound (d') may be from 0.01 to 0.5 mol, preferably from 0.02 to 0.3 mol, each being per mol of compound (a').

The thus-prepared solid catalyst component of embodiment 2 contains phthalic diester compounds (e'), (f'), and (g') described hereinabove. Of these compounds, compound (e') is the same as compound (c') and consists of one or more phthalic diesters. Compound (g') is the same as compound (d') and consists of one or more phthalic diesters.

Examples of compound (f') include n-butyl 2-ethylhexyl phthalate, isobutyl 2-ethylhexyl phthalate, methyl n-pentyl phthalate, ethyl n-pentyl phthalate, n-pentyl n-propyl phthalate, isopentyl n-propyl phthalate, n-hexyl methyl phthalate, ethyl n-hexyl phthalate, hexyl n-propyl phthalate, n-heptyl methyl phthalate, ethyl n-heptyl phthalate, n-heptyl propyl phthalate, methyl n-octyl phthalate, ethyl n-octyl phthalate, ethyl 2-ethylhexyl phthalate, n-octyl n-propyl phthalate, methyl n-nonyl phthalate, ethyl n-nonyl phthalate, n-nonyl n-propyl phthalate, n-decyl methyl phthalate, n-decyl ethyl phthalate, n-decyl n-propyl phthalate, isodecyl methyl phthalate, isodecyl ethyl phthalate, isodecyl n-propyl phthalate, methyl n-undecyl phthalate, ethyl n-undecyl phthalate, n-propyl n-undecyl phthalate, methyl n-dodecyl phthalate, ethyl n-dodecyl phthalate, and n-propyl n-dodecyl phthalate. One or more of these phthalates are contained as compound (f').

The proportions of the above-described compounds (e'), (f'), and (g') contained in the solid catalyst component of embodiment 2 are as follows. The amount of compound (e') is from 1 to 15 parts by weight, preferably from 3 to 10 parts by weight, that of compound (f') is from 0 to 10 parts by weight, preferably from 0 to 7 parts by weight, and that of compound (g') is from 0.5 to 15 parts by weight, preferably from 1 to 7 parts by weight, each being per 100 parts by weight of the solid catalyst component. The total amount of compounds (e') to (g') contained in the solid catalyst component is from 5 to 40 parts by weight, preferably from 7 to 30 parts by weight, per 100 parts by weight of the solid catalyst component.

Besides the phthalic diesters described above, the solid catalyst component of embodiment 2 further contains Mg, Ti, and a halogen as other essential ingredients. The proportions of these elements are as follows. The number of Ti atoms is from 0.01 to 0.2, preferably from 0.02 to 0.1, per atom of Mg, and the number of halogen atoms is from 10 to 100, preferably from 20 to 60, per atom of Mg. The surface area of the solid catalyst component is from 50 to 500 m<sup>2</sup>/g, preferably from 150 to 500 m<sup>2</sup>/g, and the average particle size thereof is from 3 to 150 μm, preferably from 5 to 50 μm.

Each of the solid catalyst components of embodiments 1 and 2 is also used in combination with the organoaluminum compound (B) described hereinabove and the organosilicon compound (C) represented by formula (I) to constitute the olefin polymerization catalyst of the present invention. However, in the case of using the solid catalyst component of embodiment 1 or 2, an organosilicon compound other than those represented by formula (I) may be used as component (C).

Such an organosilicon compound (referred to as "component (C1)") which can be used as component (C) in case of using the solid catalyst component (A) of embodiment 1 is a compound represented by R<sup>18</sup><sub>3</sub>Si(OR<sup>19</sup>)<sub>4-s</sub>. In the formula, R<sup>18</sup> is a straight chain or branched chain alkyl group preferably having up to 20 carbon atoms, more preferably up to 12 carbon atoms, a cycloalkyl group, an aryl group or a vinyl group, and the plurality of R<sup>18</sup> may be the same or different, R<sup>19</sup> represents a straight chain or branched chain alkyl group preferably having up to 10 carbon atoms, more preferably up to 5 carbon atoms, and s is 0, or an integer of 1 to 3. Examples of component (C1) include phenyl alkoxysilanes, alkyl alkoxysilanes, phenyl alkyl alkoxysilanes, cycloalkyl alkoxysilanes, and cycloalkyl alkyl alkoxysilanes.

To concretely exemplify the component (C1) described above, preferred are trimethyl methoxysilane, trimethyl ethoxysilane, tri-n-propyl methoxysilane, tri-n-propyl ethoxysilane, tri-n-butyl methoxysilane, tri-isobutyl methoxysilane, tri-t-butyl methoxysilane, tri-n-butyl ethoxysilane, tricyclohexyl methoxysilane, tricyclohexyl ethoxysilane, dimethyl dimethoxysilane, dimethyl diethoxysilane, di-n-propyl dimethoxysilane, di-iso-propyl dimethoxysilane, di-n-propyl diethoxysilane, di-iso-propyl diethoxysilane, di-n-butyl dimethoxysilane, di-iso-butyl dimethoxysilane, di-t-butyl dimethoxysilane, di-n-butyl diethoxysilane, n-butyl methyl dimethoxysilane, bis(2-ethylhexyl) dimethoxysilane, bis(2-ethylhexyl) diethoxysilane, dicyclohexyl dimethoxysilane, dicyclohexyl diethoxysilane, dicyclopentyl dimethoxysilane, dicyclopentyl diethoxysilane, cyclohexyl methyl dimethoxysilane, cyclohexyl methyl diethoxysilane, cyclohexyl ethyl dimethoxysilane, cyclohexyl isopropyl dimethoxysilane, cyclohexyl ethyl diethoxysilane, cyclopentyl ethyl diethoxysilane, cyclopentyl isopropyl dimethoxysilane, cyclohexyl(n-pentyl) dimethoxysilane, cyclopentyl isobutyl dimethoxysilane, cyclohexyl(n-pentyl) diethoxysilane, cyclohexyl(n-propyl) dimethoxysilane, cyclohexyl(n-butyl) dimethoxysilane, cyclohexyl(n-propyl) diethoxysilane, cyclohexyl(n-butyl) diethoxysilane, diphenyl dimethoxysilane,

diphenyl diethoxysilane, phenyl methyl dimethoxysilane, phenyl methyl diethoxysilane, phenyl ethyl dimethoxysilane, phenyl ethyl diethoxysilane, cyclohexyl dimethyl methoxysilane, cyclohexyl dimethyl ethoxysilane, cyclohexyl diethyl methoxysilane, cyclohexyl diethyl ethoxysilane, 2-ethylhexyl trimethoxysilane, 2-ethylhexyl triethoxysilane, methyl trimethoxysilane, methyl triethoxysilane, ethyl trimethoxysilane, ethyl triethoxysilane, n-propyl trimethoxysilane, n-propyl triethoxysilane, iso-propyl trimethoxysilane, iso-propyl triethoxysilane, n-butyl trimethoxysilane, iso-butyl trimethoxysilane, t-butyl trimethoxysilane, n-butyl triethoxysilane, cyclohexyl trimethoxysilane, cyclohexyl triethoxysilane, cyclopentyl trimethoxysilane, cyclopentyl triethoxysilane, vinyl trimethoxysilane, vinyl triethoxysilane, 2-ethylhexyl trimethoxysilane, 2-ethylhexyl triethoxysilane, phenyl trimethoxysilane, phenyl triethoxysilane, tetramethoxy silane and tetraethoxy silane, as well as cyclohexyl cyclopentyl dimethoxysilane. Of these, di-n-propyl dimethoxysilane, di-iso-propyl dimethoxysilane, di-n-butyl dimethoxysilane, di-iso-butyl dimethoxysilane, di-t-butyl dimethoxysilane, di-n-butyl diethoxysilane, t-butyl trimethoxysilane, dicyclohexyl dimethoxysilane, dicyclohexyl diethoxysilane, cyclohexyl dimethoxysilane, cyclohexyl methyl diethoxysilane, cyclohexyl ethyl dimethoxysilane, cyclohexyl ethyl diethoxysilane, dicyclopentyl dimethoxysilane, dicyclopentyl diethoxysilane, cyclopentyl methyl diethoxysilane, cyclopentyl ethyl diethoxysilane, and tetraethoxysilane are preferably used, as well as cyclohexyl cyclopentyl dimethoxysilane. These silicone compounds may be used independently or as admixture thereof.

Another organosilicon compound (referred to as "component (C2)") which can be used as component (C) with the solid catalyst component (A) of embodiment 2 is a compound represented by  $R^{20}_tSi(OR^{21})_{4-t}$ , wherein each  $R^{20}$  independently represents an alkyl group preferably having up to 20 carbon atoms, a cycloalkyl group preferably having 5 to 20 carbon atoms, a phenyl group, a vinyl group, an allyl group, or an aralkyl group; each  $R^{21}$  independently represents an alkyl group having 1 to 4 carbon atoms, a cycloalkyl group preferably having 5 to 20 carbon atoms, a phenyl group, an allyl group, or an aralkyl group; and t is an integer of from 1 to 3.

Examples of this organosilicon compound (C2) are as follows. Compounds wherein  $t=3$ ,  $R^{20}$  = alkyl, and  $R^{21}$  = alkyl include trimethylmethoxysilane, trimethylethoxysilane, tri-n-propylmethoxysilane, tri-n-propylethoxysilane, tri-n-butylmethoxysilane, triisobutylmethoxysilane, tri-t-butylmethoxysilane, and tri-n-butylethoxysilane. Compounds wherein  $t=3$ ,  $R^{20}$  = cycloalkyl, and  $R^{21}$  = alkyl include tricyclohexylmethoxysilane and tricyclohexylethoxysilane. Compounds wherein  $t=3$ ,  $R^{20}$  = cycloalkyl/alkyl, and  $R^{21}$  = alkyl include cyclohexyldimethylmethoxysilane, cyclohexyldimethylethoxysilane, cyclohexyldiethylmethoxysilane, cyclohexyldiethylethoxysilane, 2-ethylhexyltrimethoxysilane, and 2-ethylhexyltriethoxysilane.

Compound wherein  $t=2$ ,  $R^{20}$  = alkyl, and  $R^{21}$  = alkyl include dimethyldimethoxysilane, dimethyldiethoxysilane, di-n-propyldimethoxysilane, diisopropyldimethoxysilane, di-n-propyldiethoxysilane, diisopropyldiethoxysilane, di-n-butyldimethoxysilane, diisobutyldimethoxysilane, di-t-butyldimethoxysilane, di-n-butyl diethoxysilane, n-butylmethyldimethoxysilane, bis(2-ethylhexyl)dimethoxysilane, and bis(2-ethylhexyl)diethoxysilane. Compounds wherein  $t=2$ ,  $R^{20}$  = cycloalkyl, and  $R^{21}$  = alkyl include dicyclohexyldimethoxysilane, dicyclohexyldiethoxysilane, dicyclopentyldimethoxysilane, dicyclopentyldiethoxysilane, and cyclohexylcyclopentyldimethoxysilane. Compounds wherein  $t=2$ ,  $R^{20}$  = cycloalkyl/alkyl, and  $R^{21}$  = alkyl include cyclohexylmethylmethoxysilane, cyclohexylmethyldiethoxysilane, cyclohexylethyldimethoxysilane, cyclohexylethylethoxysilane, cyclohexylisopropyldimethoxysilane, cyclohexylethyldiethoxysilane, cyclopentylethyldiethoxysilane, cyclopentylisopropyldimethoxysilane, cyclohexyl-n-pentyldimethoxysilane, cyclopentylisobutyldimethoxysilane, cyclohexyl-n-pentyldiethoxysilane, cyclohexyl-n-propyldimethoxysilane, cyclohexyl-n-butyldimethoxysilane, cyclohexyl-n-propyldiethoxysilane, and cyclohexyl-n-butyldiethoxysilane. Compounds wherein  $t=2$ ,  $R^{20}$  = phenyl, and  $R^{21}$  = alkyl include diphenyl dimethoxysilane and diphenyldiethoxysilane. Compounds wherein  $t=2$ ,  $R^{20}$  = phenyl or alkyl, and  $R^{21}$  = alkyl include phenylmethyldimethoxysilane, phenylmethyldiethoxysilane, phenylethyldimethoxysilane, and phenylethyldiethoxysilane.

Compounds wherein  $t=1$ ,  $R^{20}$  = alkyl, and  $R^{21}$  = alkyl include methyltrimethoxysilane, methyltriethoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, n-propyltrimethoxysilane, n-propyltriethoxysilane, isopropyltrimethoxysilane, isopropyltriethoxysilane, n-butyltrimethoxysilane, isobutyltrimethoxysilane, t-butyltrimethoxysilane, n-butyltriethoxysilane, 2-ethylhexyltrimethoxysilane, and 2-ethylhexyltriethoxysilane. Compounds wherein  $t=1$ ,  $R^{20}$  = cycloalkyl, and  $R^{21}$  = alkyl include cyclohexyltrimethoxysilane, cyclohexyltriethoxysilane, cyclopentyltrimethoxysilane, and cyclopentyltriethoxysilane. Compounds wherein  $t=1$ ,  $R^{20}$  = vinyl, and  $R^{21}$  = alkyl include vinyltrimethoxysilane and vinyltriethoxysilane. Further, compounds wherein  $t=1$ ,  $R^{20}$  = phenyl, and  $R^{21}$  = alkyl include phenyltrimethoxysilane and phenyltriethoxysilane.

Preferred of the above organosilicon compounds (C2) are the compounds wherein  $t=2$ ,  $R^{20}$  =  $C_1$ - $C_4$  alkyl, and  $R^{21}$  =  $C_1$ - $C_2$  alkyl, the compounds wherein  $t=2$ ,  $R^{20}$  = cycloalkyl, and  $R^{21}$  =  $C_1$ - $C_2$  alkyl, the compounds wherein  $t=2$ ,  $R^{20}$  = cycloalkyl or  $C_1$ - $C_2$  alkyl, and  $R^{21}$  =  $C_1$ - $C_2$  alkyl, and the compounds wherein  $t=1$ ,  $R^{20}$  =  $C_3$ - $C_4$  neoalkyl, and  $R^{21}$  =  $C_1$ - $C_2$  alkyl. These compounds, when used in combination with the

solid catalyst component (A) of embodiment 2, effectively function to produce a polymer having an especially wide molecular weight distribution and high crystallinity in high yield.

Specific examples of such preferred organosilicon compounds include di-n-propyldimethoxysilane, diisopropyldimethoxysilane, di-n-butyldimethoxysilane, diisobutyldimethoxysilane, di-t-butyldimethoxysilane, di-n-butyldiethoxysilane, t-butyltrimethoxysilane, dicyclohexyldimethoxysilane, dicyclohexyldiethoxysilane, cyclohexyldimethoxysilane, cyclohexylmethyldiethoxysilane, cyclohexylethyldimethoxysilane, cyclohexylethyldiethoxysilane, dicyclopentyldimethoxysilane, dicyclopentyldiethoxysilane, cyclopentylmethyldiethoxysilane, and cyclopentylethyldiethoxysilane, as well as cyclohexylcyclopentyldimethoxysilane. Especially preferred organosilicon compounds of these are t-butyltrimethoxysilane, dicyclohexyldimethoxysilane, dicyclopentyldimethoxysilane, n-butylmethyldimethoxysilane, cyclohexylisopropyldimethoxysilane, cyclopentylisopropyldimethoxysilane, and cyclopentylisobutyldimethoxysilane, as well as cyclohexylcyclopentyldimethoxysilane. These organosilicon compounds may be used alone or in combination of two or more thereof.

It is preferred to wash the thus prepared solid catalyst component (A) of embodiment 1 or 2 in the inert organic solvent such as heptane, whereby unreacted substances can be removed. With or without drying, the washed component (A) of embodiment 1 or 2 is mixed with the organic aluminium compound (B) and the silicone compound (C1) or (C2) as described in detail above, to thereby form the catalyst for polymerizing olefins of the present invention.

In preparation of the polymerization catalyst using the solid catalyst component (A) of embodiment 1, the component (B) is used in an amount of 1 to 1000 moles, preferably 50 to 500 moles per mole of the titanium atom contained in the solid catalyst component (A), and the component (C1) is used in an amount of 0.0020 to 2 moles, preferably 0.01 to 0.5 mole per mole of the component (B). The solid catalyst component (A) of embodiment 1 used in the catalyst of the present invention generally has a Ti content of 0.5 to 15% by weight and preferably 1 to 7% by weight.

In the case of using the solid catalyst component (A) of embodiment 2, the amounts of components (B) and (C2) are from 5 to 1,000 mol and from 0.002 to 0.5 mol, respectively, per mol of the titanium atom contained the solid catalyst component (A).

Polymerization reaction according to the present invention may be carried out in the presence or absence of an organic solvent. The olefin monomer to be polymerized may be used in either a gaseous state or a liquid state. The polymerization is conducted at a temperature of not higher than 200°C, preferably not higher than 100°C, under a pressure of not higher than 10 MPa, preferably not higher than 5 MPa. The reaction may be effected either in a continuous system or in a batch system and through one step or two or more steps.

The olefins to be homo- or copolymerized according to the present invention are not particularly limited and generally have 2 to 20 carbon atoms, such as ethylene, propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, and vinylcyclohexane. These olefins may be used either individually or in combination of two or more thereof. The effects of the present invention in assuring high stereoregularity, broad molecular weight distribution, and high yield are particularly pronounced in homopolymerization of propylene or copolymerization of propylene and ethylene.

For ensuring the improvements in catalytic activity and stereoregularity and particle properties of the polymer produced, it is preferable to conduct pre-polymerization prior to substantial polymerization. Monomers to be pre-polymerized include not only ethylene and propylene but other monomers, such as styrene and vinylcyclohexane.

The catalyst of the present invention is used in an amount of about 0.005 to 0.5 mmol, preferably about 0.01 to 0.5 mmol, calculated as titanium atom in solid catalyst component (A) per liter of the polymerization zone.

According to the process of the present invention, the olefin polymers obtained have a broader molecular weight distribution than those obtained by conventional processes, by at least 1 higher as expressed in terms of the ratio of weight average molecular weight to number average molecular weight (Mw/Mn) of the olefin polymers, and the yield of stereoregular polymers is extremely high. That is, the process has been confirmed to provide polyolefins having not only broad molecular weight distribution (for example, 6 or higher in terms of the Mw/Mn) but high stereoregularity in extremely high yield.

The present invention will now be illustrated in greater detail with reference to Examples in view of Comparative Examples, but it should be understood that the present invention is not construed as being limited to these Examples. All the percents are by weight unless otherwise indicated.

**EXAMPLE 1**

Into a 2-liter four-necked flask equipped with a stirrer, thermometer, Dimroth condenser, and dropping funnel was introduced 18.5 g (0.76 mol) of magnesium shavings. The magnesium was dried in an argon stream, and 20 ml of di-n-butyl ether was then added thereto. The contents were cooled to room temperature, and a small amount of 1,2-dibromoethane was added thereto to activate the magnesium. A solution prepared by dissolving 79.6 g (0.76 mol) of cyclopentyl chloride in 600 ml of di-n-butyl ether was then added dropwise over a period of 3.5 hours, during which the temperature of the system spontaneously increased to 50°C. Subsequently, 143.0 g (0.70 mol) of cyclohexyltrimethoxysilane was added thereto at room temperature, and the reaction was then conducted for 1 hour under reflux.

After completion of the reaction, the reaction mixture was cooled to room temperature, and 372 g (0.38 mol) of a 10% aqueous sulfuric acid solution was added thereto dropwise at a temperature of 40°C or lower. The organic layer was washed with 300 ml of a 1% aqueous sodium hydrogen carbonate solution and then dried over anhydrous magnesium sulfate. After the drying agent was filtered off, vacuum distillation was performed to obtain 143.6 g of a fraction having a boiling point of 78°C at 0.2 Torr. The yield was 84.6%. This reaction product was ascertained to be cyclohexylcyclopentyltrimethoxysilane by MS, two-dimensional analysis with <sup>1</sup>H-NMR/<sup>13</sup>C-NMR, and IR. The results of MS, <sup>1</sup>H-NMR/<sup>13</sup>C-NMR (COSY spectrum), and IR are shown in Figs. 1, 2, and 3, respectively.

The analyses by MS, <sup>1</sup>H-NMR/<sup>13</sup>C-NMR, and IR were carried out under the following conditions.

MS: apparatus ... Finigan Mat (GC-MS).  
<sup>1</sup>H-NMR/<sup>13</sup>C-NMR: apparatus ... JEOL GSX270,  
 solvent ... CDCl<sub>3</sub>.  
 IR: apparatus ... Perkin Elmer 1600 Series (FT-IR),  
 KBr sand method.

**EXAMPLE 2****Preparation of Solid Catalyst Component:**

Into a 200-ml round-bottom flask the inside atmosphere of which had been sufficiently replaced with nitrogen gas and which was equipped with a stirrer were introduced 10 g of diethoxymagnesium and 80 ml of toluene. The contents were stirred to obtain a suspension. To this suspension was added 20 ml of titanium tetrachloride. The mixture was heated and, at the time when the temperature thereof had reached 62°C, 1.0 ml of diethyl phthalate was added. This mixture was then heated and, at the time when the temperature thereof had reached 110°C, 3.5 ml of dioctyl phthalate was added. The resulting mixture was heated to 112°C and stirred at this temperature for 1.5 hours to allow a reaction to proceed. After completion of the reaction, the reaction product was washed twice with 100 ml of toluene heated at 90°C. To the washed reaction product were added 20 ml of titanium tetrachloride and 80 ml of toluene. This mixture was heated to 100°C and stirred for 2 hours to allow a reaction to proceed. After completion of the reaction, the reaction product was washed 10 times with 100 ml of n-heptane warmed at 40°C to obtain a solid catalyst component. The titanium content of this solid catalyst component was measured and found to be 2.46% by weight.

**Formation of Polymerization Catalyst and Polymerization of Olefin:**

Into a 2.0-liter autoclave the inside atmosphere of which had been sufficiently replaced with nitrogen gas and which was equipped with a stirrer were introduced 1.32 mmol of triethylaluminum, 0.13 mmol of cyclohexylcyclopentyltrimethoxysilane, and 0.0066 mmol of the solid catalyst component in terms of the amount of titanium atoms. Thus, a polymerization catalyst was formed. Thereafter, 1.8 liters of hydrogen gas and 1.4 liters of liquefied propylene were introduced into the autoclave to conduct polymerization at 70°C for 30 minutes. The weight (a) of the polymer thus obtained was 349.1 g. When this polymer was extracted with boiled n-heptane for 6 hours, the n-heptane-insoluble amount (b) was 343.2 g. The catalytic activity was 23,900 g per g of the solid catalyst component used. The yield of the wholly crystalline polymer was 98.3%. The polymer yielded had an MI of 3.6 g/10-min, a molecular weight distribution of 7.1, and a melting point of 164.0°C.

The catalytic activity per unit weight of the solid catalyst component used was calculated using the following equation.

$$\text{Catalytic Activity} = \frac{(a) (g)}{\text{Solid Catalyst Component (g)}}$$

The yield of wholly crystalline polymer was calculated using the following equation.

$$\text{Yield of Wholly Crystalline Polymer} = \frac{(b)}{(a)} \times 100 (\%)$$

The molecular weight distribution was determined using the following equation.

$$\text{Molecular Weight Distribution} = \frac{\text{Weight-average Molecular Weight (M}_w\text{)}}{\text{Number-average Molecular Weight (M}_n\text{)}}$$

As described above, the organosilicon compound of the present invention, when used as an electron donor serving as one component of an olefin polymerization catalyst, gives a polyolefin having a broad molecular weight distribution and high crystallinity, while retaining high performances with respect to catalytic activity and the yield of highly stereoregular polymer which performances are equal to or higher than those of conventionally known high-performance catalysts.

### EXAMPLE 3

#### Preparation of Solid Catalyst Component:

In a 200 ml-volume round flask equipped with a stirrer having been thoroughly purged with nitrogen gas were charged 10 g of diethoxymagnesium and 80 ml of toluene to prepare a suspension. To the suspension was added 20 ml of titanium tetrachloride, the mixture was heated to 80°C, at which 2.7 ml of di-n-butyl phthalate was added. The mixture was further heated up to 110°C, at which the mixture was allowed to react for 2 hours with stirring. After completion of the reaction, the reaction mixture was washed with two 100 ml portions of toluene at 90°C, and 20 ml of titanium tetrachloride and 80 ml of toluene were added thereto. The mixture was heated to 100°C, at which it was allowed to react for 2 hours while stirring. After completion of the reaction, the reaction mixture was washed with ten 100 ml portions of n-heptane at 40°C to obtain a solid catalyst component. The solid content of the solid catalyst component, separated by solid-liquid separation, was found to have a titanium content of 2.91%.

#### Preparation of Catalyst System and Polymerization:

In the same manner as in Example 2, a catalyst system was prepared using the above-prepared solid catalyst component and propylene was polymerized in the presence of the catalyst system. The properties of the resulting polymer are shown in Table 5 below.

### EXAMPLE 4

#### Preparation of Solid Catalyst Component:

In a 200 ml-volume round flask equipped with a stirrer having been thoroughly purged with nitrogen gas were charged 20 ml of titanium tetrachloride and 30 ml of toluene to prepare a mixed solution. To the mixed solution was added a suspension of 10 g of spherical diethoxymagnesium particles, 50 ml of

toluene, and 3.6 ml of di-n-butyl phthalate, and the mixture was heated to 90 °C, at which it was allowed to react for 1 hour with stirring. After completion of the reaction, the reaction mixture was washed with two 100 ml portions of toluene at 90 °C, and 20 ml of titanium tetrachloride and 80 ml of toluene were added thereto. The mixture was heated to 110 °C, at which it was allowed to react for 2 hours while stirring. After completion of the reaction, the reaction mixture was washed ten 100 ml portions of n-heptane at 40 °C to obtain a solid catalyst component. The solid content of the solid catalyst component, separated by solid-liquid separation, was found to have a titanium content of 2.87%.

#### Preparation of Catalyst System and Polymerization:

Propylene was polymerized in the same manner as in Example 2, except for using the above-prepared solid catalyst component. The reaction results are shown in Table 5.

#### EXAMPLE 5

##### Preparation of Solid Catalyst Component:

In a 200 ml-volume round flask equipped with a stirrer having been thoroughly purged with nitrogen gas were charged 10 g of diethoxymagnesium and 80 ml of toluene to prepare a suspension. To the suspension was added 20 ml of titanium tetrachloride, and the mixture was heated to 60 °C, at which 1.0 ml of diethyl phthalate was added. The mixture was further heated up to 110 °C, at which 2.5 ml of di-isooctyl phthalate was added thereto. The mixture was further heated to 112 °C, at which the mixture was allowed to react for 1.5 hours with stirring. After completion of the reaction, the reaction mixture was washed with two 100 ml portions of toluene at 90 °C, and 20 ml of titanium tetrachloride and 80 ml of toluene were added thereto. The mixture was heated to 100 °C, and it was allowed to react at that temperature for 2 hours while stirring. After completion of the reaction, the reaction mixture was washed with ten 100 ml portions of n-heptane at 40 °C to obtain a solid catalyst component. The solid content of the solid catalyst component, separated by solid-liquid separation, was found to have a titanium content of 2.74%.

##### Preparation of Catalyst System and Polymerization:

Propylene was polymerized in the same manner as in Example 2, except for using the above-prepared solid catalyst component. The reaction results are shown in Table 5.

#### EXAMPLE 6

##### Preparation of Solid Catalyst Component:

In a 200 ml-volume round flask equipped with a stirrer having been thoroughly purged with nitrogen gas were charged 10 g of diethoxymagnesium and 80 ml of toluene to prepare a suspension. To the suspension was added 20 ml of titanium tetrachloride, and the mixture was heated to 62 °C, at which 1.0 ml of diethyl phthalate was added. The mixture was heated up to 110 °C, at which 4.0 ml of di-isooctyl phthalate was added thereto. The mixture was further heated to 112 °C, at which the mixture was allowed to react for 1.5 hours with stirring. After completion of the reaction, the reaction mixture was washed with two 100 ml portions of toluene at 90 °C, and 20 ml of titanium tetrachloride and 80 ml of toluene were added thereto. The mixture was heated to 100 °C, and it was allowed to react at that temperature for 2 hours while stirring. After completion of the reaction, the reaction mixture was washed with ten 100 ml portions of n-heptane at 40 °C to obtain a solid catalyst component. The solid content of the solid catalyst component, separated by solid-liquid separation, was found to have a titanium content of 2.17%.

##### Preparation of Catalyst System and Polymerization:

Propylene was polymerized in the same manner as in Example 2, except for using the above-prepared solid catalyst component. The reaction results are shown in Table 5.

COMPARATIVE EXAMPLE 1

Propylene was polymerized in the same manner as in Example 3, except for replacing cyclohexylcyclopentylmethoxysilane with phenyltriethoxysilane. The reaction results are shown in Table 5.

COMPARATIVE EXAMPLE 2

Propylene was polymerized in the same manner as in Example 3, except for replacing cyclohexylcyclopentylmethoxysilane with cyclohexylmethyldimethoxysilane. The reaction results are shown in Table 5.

COMPARATIVE EXAMPLE 3

Propylene was polymerized in the same manner as in Example 3, except for replacing cyclohexylcyclopentylmethoxysilane with dicyclopentylmethoxysilane. The reaction results are shown in Table 5.

EXAMPLE 7Preparation of Solid Catalyst Component:

In a 200 ml-volume round flask equipped with a stirrer having been thoroughly purged with nitrogen gas were charged 7.14 g of anhydrous magnesium chloride, 37.5 ml of decane and 35.1 ml of 2-ethylhexyl alcohol, and the resulting mixture was heated at 130 °C for 2 hours to obtain a uniform solution. Then 1.67 g of phthalic anhydride was added thereto, followed by stirring at 130 °C for one hour. The thus obtained uniform solution was cooled to room temperature and was dropwise added, over one hour, to 200 ml of titanium tetrachloride which had been cooled at -20 °C. After addition of the solution, the temperature of the resulting solution was increased to 110 °C over 4 hours, at which 5.03 ml of diisobutyl terephthalate was further added to the solution, and the resulting solution was then stirred for 2 hours to continue the reaction at 110 °C. The hot reaction mixture was subjected to filtration to thereby obtain a solid product which was then dispersed in 275 ml of titanium tetrachloride and allowed to stand at 110 °C for 2 hours. Thereafter, a solid product was separated again from the dispersion by filtration while the dispersion was hot, and the solid product was washed with decane and heptane at 110 °C to obtain a solid catalyst component. The solid content of the solid catalyst component, separated by solid-liquid separation, was found to have a titanium content of 2.06 %.

Preparation of Catalyst System and Polymerization:

Propylene was polymerized in the same manner as in Example 2, except for using the above-prepared solid catalyst component. The reaction results are shown in Table 5.

EXAMPLE 8Preparation of Solid Catalyst Component:

In a 250 ml-volume round flask equipped with a stirrer having been thoroughly purged with nitrogen gas was charged a solution of 1.4 ml of titanium tetrachloride dissolved in 74 ml of chlorobenzene, and 3.6 ml of diisobutyl phthalate and 11.8 g of diethoxymagnesium were subsequently added thereto. To the resulting solution was further added a solution of 94 ml of titanium tetrachloride dissolved in 24 ml of chlorobenzene. The addition of these compounds and the solution was conducted at a temperature of 20 to 25 °C. The resulting mixture was heated at 110 °C with stirring for one hour, followed by filtration while the mixture was hot. The thus obtained solid product was added to a solution of 94 ml of titanium tetrachloride dissolved in 24 ml of chlorobenzene to form a slurry at room temperature. Then, a solution obtained by dissolving 0.9 g of phthaloyl dichloride in 74 ml of chlorobenzene was added to the slurry at room temperature, followed by heating at 110 °C with stirring for 30 minutes. The resulting mixture was filtered while it was hot, whereby a solid product was obtained.

To the thus obtained solid product was added, at room temperature, a solution of 94 ml of titanium tetrachloride dissolved in 24 ml of chlorobenzene to thereby form a slurry. 74 ml of chlorobenzene was further added, at room temperature, to the slurry which was then heated at 110 °C with stirring for 30

minutes. The resulting mixture was filtered while it was hot, whereby a solid product was obtained. Using the thus obtained solid product, the above procedure was repeated again to obtain a solid product, which was then washed 10 times with 100 ml of heptane at 25 °C. Thus, a solid catalyst component was obtained. The solid content thereof, separated by solid-liquid separation, was found to have a titanium content of 2.63 %.

Preparation of Catalyst system and Polymerization:

Propylene was polymerized in the same manner as in Example 2, except for using the above-prepared solid catalyst component. The reaction results are shown in Table 5.

COMPARATIVE EXAMPLE 4

Propylene was polymerized in the same manner as in Example 7, except for replacing cyclohexylcyclopentylmethoxysilane with cyclohexylmethyldimethoxysilane. The reaction results are shown in Table 5.

COMPARATIVE EXAMPLE 5

Propylene was polymerized in the same manner as in Example 8, except for replacing cyclohexylcyclopentylmethoxysilane with cyclohexylmethyldimethoxysilane. The reaction results are shown in Table 5.

TABLE 5

Example No.	Weight of Polymer Produced (g)	n-Heptane Insoluble Content (g)	Polymerization Activity (g/g-cat.)	Yield of Total Crystalline Polymer (%)	MI (g/10 min)	Molecular Weight Distribution
Example 3	306.4	302.3	28,200	98.7	5.0	7.0
Example 4	339.5	334.4	30,800	98.5	3.0	6.5
Example 5	293.1	289.0	25,500	98.6	3.6	7.1
Example 6	349.1	343.2	23,900	98.3	3.6	7.1
Example 7	177.1	173.9	23,100	98.2	4.5	6.5
Example 8	217.1	215.6	36,200	99.3	3.7	6.4
Comp. Example 1	220.7	217.6	20,300	98.6	10.0	5.2
Comp. Example 2	322.7	317.5	29,700	98.4	5.0	5.4
Comp. Example 3	385.7	380.7	35,500	98.7	2.5	4.9
Comp. Example 4	192.3	188.8	25,100	98.2	5.2	5.2
Comp. Example 5	215.9	211.4	36,000	97.9	5.0	5.1

It is seen from the Examples that polymerization of an olefin in the presence of the catalyst comprising the asymmetric organosilicon compound containing a cyclohexyl group and a cyclopentyl group of the present invention provides an olefin polymer having high stereoregularity and broad molecular weight distribution in high yield.

**EXAMPLE 9**Preparation of Solid Catalyst Component:

5 10 g of diethoxymagnesium, 1.5 g of aluminium trichloride and 90 ml of toluene were put in a 500-ml round flask equipped with a stirrer, which had been fully purged with nitrogen gas, and they were mixed to prepare a suspension. Then, 22 ml of titanium tetrachloride (maintained at a room temperature) was added therein, and the temperature was increased to 80 °C while stirring to carry out a reaction. Next, 3.0 ml of di-n-butyl phthalate was added, and the temperature was further increased to 110 °C to continue the reaction  
 10 for 2 hours. Thereafter, a supernatant of the resulting suspension was removed, and the residue was washed three times with 88 ml of toluene at 75 °C. Subsequently, 89 ml of toluene and 22 ml of titanium tetrachloride were added thereto, and processing was carried out at 100 °C for 1.5 hour while stirring, followed by washing eight times with 83 ml of heptane at 40 °C, whereby a solid catalyst component having a Ti content of 3.8 % by weight and an Al content of 0.5 % by weight was obtained.

Preparation of Catalyst and Polymerization of Propylene:

700 ml of n-heptane was put in a 1800-ml stainless steel autoclave equipped with a stirrer, which had been fully dried with nitrogen gas and replaced with propylene gas. Under the propylene gas atmosphere ,  
 20 2.10 mmol of triethylaluminium, 0.21 mmol of phenyl triethoxysilane and 0.084 mmol (in terms of Ti) of the above-prepared solid catalyst component were put therein in the autoclave, whereby a catalyst was prepared. Thereafter, a propylene pressure was adjusted to 0.2 MPa, and preliminary polymerization of propylene was carried out at 20 °C for 30 minutes with stirring. 30 ml of hydrogen gas was then introduced in the autoclave, and the propylene pressure in the system was adjusted to 0.7 MPa to continue the  
 25 polymerization at 70 °C for 2 hours. The pressure, which decreased as the polymerization of propylene proceeded, was supplemented by continuously supplying only propylene to keep the pressure constant during the polymerization. The polymer thus produced was filtered off and dried under reduced pressure to obtain a solid polymer.

Measurement of Properties of Catalyst:

A filtrate separated from the solid polymer was concentrated to obtain the polymer dissolved in the solvent, and the amount of the polymer was designated as (A), while the amount of the solid polymer was designated as (B). Further, the solid polymer obtained was extracted in boiled n-heptane for 6 hours to  
 35 obtain a polymer insoluble in n-heptane, and the amount thereof was designated as (C). The properties of the catalyst were calculated from these values (A), (B) and (C), as described below.

Polymerization activity (Y) per unit amount of solid catalyst component:

$$Y = \frac{[(A) + (B)](g)}{\text{amount of solid catalyst component (g)}}$$

$$(RDS) = \frac{(A)(g)}{[(A) + (B)](g)}$$

40 Yield (t-II) of a whole crystalline polymer:

$$(t-II) = \frac{(C)(g)}{[(A) + (B)](g)}$$

Further, a density ( $\rho$ ), a melt index (MI) (measured by the method according to JIS K7210) and a bulk density (BD) of the resulting solid polymer were measured, and the results are shown in Table 6, together with the above-measured properties of the catalyst.

**EXAMPLE 10**Preparation of Solid Catalyst Component:

50 10 g of diethoxymagnesium and 90 ml of toluene were put in a 500-ml round flask equipped with a stirrer, which had been fully replaced with nitrogen gas, to prepare a suspension. Then, 20 ml of titanium tetrachloride was added therein, and the temperature was increased to 70 °C while stirring to carry out the reaction. Next, 4.0 ml of di-i-octyl phthalate was added, and the temperature in the system was further increased to 110 °C to continue the reaction for 2 hours. After completion of the reaction, a supernatant of  
 55 the resulting suspension was removed, and the residue was washed three times with 88 ml of toluene at 75 °C. Thereafter, 80 ml of toluene, 2.0 g of aluminium trichloride and 30 ml of titanium tetrachloride were added, and the reaction was carried out at 105 °C for 2 hours with stirring, followed by washing eight times with 80 ml of heptane at 40 °C, whereby a solid catalyst component having a Ti content of 3.4 % by weight

and an Al content of 0.7 % by weight was obtained.

#### Preparation of Catalyst and Polymerization of Propylene:

- 5 Propylene was polymerized in the same manner as that in Example 9, except that phenyl triethoxysilane was replaced with dicyclohexyl dimethoxysilane, and the results are shown in Table 6.

#### EXAMPLE 11

##### 10 Preparation of Solid Catalyst Component:

- 10 g of diethoxymagnesium, 0.8 g of aluminium trichloride and 90 ml of toluene were put in a 500-ml round flask equipped with a stirrer, which has been fully purged with nitrogen gas, to prepare a suspension. Then, 22 ml of titanium tetrachloride was added therein, and the temperature was increased to 80 °C while stirring to carry out the reaction. Next, 2.8 ml of di-*i*-octyl phthalate was added, and the temperature was further increased to 110 °C to continue the reaction for 2 hours. After completion of the reaction, a supernatant of the resulting suspension was removed, and the residue was washed twice with 88 ml of toluene at 75 °C. Thereafter, 0.8 g of aluminium trichloride, 89 ml of toluene and 22 ml of titanium tetrachloride were added, and the processing was carried out at 100 °C for 1.5 hour while stirring, followed by washing eight times with 83 ml of heptane at 40 °C, whereby a solid catalyst component having a Ti content of 2.9 % by weight and an Al content of 0.8 % by weight was obtained.

#### Preparation of Catalyst and Polymerization of Propylene:

- 25 Propylene was polymerized in the same manner as that in Example 9, except that phenyl triethoxysilane was replaced with cyclohexyl methyl dimethoxysilane, and the results are shown in Table 6.

#### COMPARATIVE EXAMPLE 6

- 30 A solid catalyst component was prepared and polymerization of propylene was carried out in the same manner as in Example 9, except that aluminium trichloride was not used. The results obtained are shown in Table 6.

#### COMPARATIVE EXAMPLE 7

- 35 A solid catalyst component was prepared in the same manner as in Example 9, except that aluminium trichloride was not used and the amount of di-*n*-butyl phthalate was changed to 2.0 ml. The resulting solid catalyst component had a Ti content of 5.5 % by weight.

- 40 Using the solid catalyst component, polymerization of propylene was carried out in the same manner as in Example 9, and the results are shown in Table 6.

TABLE 6

	Ex.9	Ex.10	Ex.11	Comp.Ex.6	Comp.Ex.7
45 Polymerization activity (Y)	11,500	13,000	26,000	11,700	10,700
RDS*	1.0	0.9	0.8	0.5	4.5
Yield of whole crystalline polymer (t-II)	96.1	95.4	95.5	98.2	92.6
Density of polymer ( $\rho$ : g/ml)	0.9052	0.9048	0.9045	0.9080	0.9049
50 Melt index of polymer (MI: g/10 min)	3.7	3.5	1.9	2.2	4.0
Bulk density of polymer (BD: g/ml)	0.41	0.40	0.39	0.40	0.38

(\* in *n*-heptane)

- 55 It is seen from the results shown in Table 6 that the use of the solid catalyst component of the present invention produces polypropylene having a density of not more than 0.906 g/ml at a low RDS of 3% or less.

EXAMPLE 12Preparation of Solid Catalyst Component:

5 10 g of diethoxymagnesium, 1.5 g of triethoxyaluminium and 90 ml of toluene were put in a 500-ml round flask equipped with a stirrer, which had been fully purged with nitrogen gas, to prepare a suspension. Then, 30 ml of titanium tetrachloride was added therein, and the temperature was increased to 80 °C while stirring to carry out the reaction. Next, 3.0 ml of di-n-butyl phthalate was added, and the temperature was further increased to 110 °C to continue the reaction for 2 hours. After completion of the reaction, a  
 10 supernatant of the resulting suspension was removed, and the residue was washed three times with 90 ml of toluene at 75 °C. Thereafter, 90 ml of toluene and 30 ml of titanium tetrachloride were added, and the processing was carried out at 100 °C for 2 hours while stirring, followed by washing eight times with 80 ml of heptane at 40 °C, whereby a solid catalyst component having a Ti content of 5.5 % by weight and an Al content of 0.8 % by weight was obtained.

Preparation of Catalyst and Polymerization of Propylene:

Propylene was polymerized in the same manner as that in Example 9, except that phenyl triethoxysilane was replaced with cyclohexyl methyl dimethoxysilane. The results are shown in Table 7.

EXAMPLE 13

A solid catalyst component was prepared in the same manner as that in Example 12, except that triethoxyaluminium was replaced with triisopropoxyaluminium. Using the solid catalyst component, polymerization of propylene was carried out in the same manner as in Example 12, and the results are shown in Table 7.

EXAMPLE 14

A solid catalyst component was prepared in the same manner as that in Example 12, except that triethoxyaluminium was replaced with ethoxydichloroaluminium. Using the solid catalyst component polymerization of propylene was carried out in the same manner as in Example 12, and the results are shown in Table 7.

EXAMPLE 15Preparation of Solid Catalyst Components:

10 g of diethoxymagnesium, 1.5 g of triisopropoxyaluminium and 90 ml of toluene were put in a 500-ml  
 40 round flask equipped with a stirrer, which had been fully purged with nitrogen gas, to prepare a suspension. 30 ml of titanium tetrachloride was added therein, and the temperature was increased to 80 °C with stirring to carry out the reaction. Next, 3.5 ml of di-i-octyl phthalate was added, and the temperature was further increased 110 °C to continue the reaction for 2 hours. After completion of the reaction, a supernatant of the resulting suspension was removed, and the residue was washed with 90 ml of toluene at 75 °C. Thereafter,  
 45 0.5 g of ethoxydichloroaluminium, 90 ml of toluene and 30 ml of titanium tetrachloride were added, and the processing was carried out at 100 °C for 2 hours while stirring, followed by washing eight times with 80 ml of heptane at 40 °C, whereby a solid catalyst component having a Ti content of 4.2 % by weight and an Al content of 0.8 % by weight was obtained.

Preparation of Catalyst and Polymerization of Propylene:

Propylene was polymerized in the same manner as that in Example 12, except that 0.0053 mmol in terms of Ti of the above-prepared solid catalyst component Ti was used, and the results are shown in Table 7.

## COMPARATIVE EXAMPLE 8

A solid catalyst component was prepared and polymerization of propylene was carried out in the same manner as in Example 12, except that triethoxyaluminium was not used. The results obtained are shown in Table 7.

TABLE 7

	Ex.12	Ex.13	Ex.14	Ex.15	Comp. Ex.8
Polymerization activity (Y)	24,200	26,100	28,900	25,700	24,500
RDS*	1.5	1.2	1.0	1.4	0.5
Yield of whole crystalline polymer (t-II)	95.0	95.3	95.4	94.8	98.5
Density of polymer ( $\rho$ : g/ml)	0.9050	0.9052	0.9048	0.9047	0.9078
Melt index of polymer (MI: g/10 min)	4.0	2.8	1.8	1.5	1.8
Bulk density of polymer (BD: g/ml)	0.40	0.41	0.40	0.40	0.40
Ti content (wt%)	5.5	5.2	3.5	4.2	2.5
Al content (wt%)	0.8	0.7	0.6	0.8	-
(* in n-heptane)					

## EXAMPLE 16

## Preparation of Solid Catalyst Component:

10 g of diethoxymagnesium, 1.5 g of aluminium trichloride and 90 ml of toluene were put in a 500-ml round flask equipped with a stirrer, which had been fully purged with nitrogen gas, to prepare a suspension. Then, 22 ml of titanium tetrachloride was added therein, and the temperature was increased to 80°C while stirring to carry out the reaction. Next, 3.3 ml of di-n-butyl phthalate and 3.0 ml of dimethyl polysiloxane having a viscosity of 50 cst at room temperature were added, and the temperature was further increased to 110°C to continue the reaction for 2 hours. After completion of the reaction, a supernatant of the resulting suspension was removed and the residue was washed three times with 88 ml of toluene at 75°C. Thereafter, 89 ml of toluene and 22 ml of titanium tetrachloride were added, and the processing was carried out at 100°C for 1.5 hour while stirring, followed by washing eight times with 83 ml of heptane at 40°C, whereby a solid catalyst component having a Ti content of 3.3 % by weight and an Al content of 0.5 % by weight was obtained.

## Preparation of Catalyst and Polymerization of Propylene:

Using the above-prepared solid catalyst component, polymerization of propylene was carried out in the same manner as in Example 15, and the results are shown in Table 8.

## EXAMPLE 17

## Preparation of Solid Catalyst Components:

10 g of diethoxymagnesium, 1.0 g of aluminium trichloride and 90 ml of toluene were put in a 500-ml round flask equipped with a stirrer, which had been fully purged with nitrogen gas, to prepare a suspension. Then, 20 ml of titanium tetrachloride was added therein, and the temperature was increased 50°C while stirring to carry out the reaction. Next, 4.5 ml of di-i-octyl phthalate was added, and the temperature was added to continue the reaction for 2 hours. After completion of the reaction, a supernatant of the resulting suspension was removed, and the residue was washed three times with 88 ml of toluene at 75°C. Thereafter, 80 ml of toluene, 1.0 g of aluminium chloride and 30 ml of titanium tetrachloride were added, and the reaction was carried out at 105°C for 2 hours while stirring, followed by washing eight times with 80 ml of heptane at 40°C, whereby a solid catalyst component having a Ti content of 2.9 % by weight and an Al content of 0.8 % by weight was obtained.

Preparation of Catalyst and Polymerization of Propylene:

Propylene was polymerized in the same manner as in Example 15, except that cyclohexyl methyl dimethoxysilane was replaced with diphenyl dimethoxysilane, and the results are shown in Table 8.

EXAMPLE 18Preparation of Solid Catalyst Component:

10 g of diethoxymagnesium, 0.8 g of aluminium trichloride and 90 ml of toluene were put in a 500-ml round flask equipped with a stirrer, which had been fully purged with nitrogen gas, to prepare a suspension. Thus, 22 ml of titanium tetrachloride was added therein, and the temperature was increased to 80 °C while stirring to carry out the reaction. Next, 4.8 ml of di-i-octyl phthalate was added, and the temperature was further increased to 110 °C, and then 6.0 ml of dimethyl polysiloxane having a viscosity of 100 cst at room temperature was added to continue the reaction for 2 hours. After completion of the reaction, a supernatant of the resulting suspension was removed, and the residue was washed twice with 88 ml of toluene at 75 °C. Thereafter, 0.8 g of aluminium chloride, 89 ml of toluene and 22 ml of titanium tetrachloride were added, and the processing was carried out at 100 °C for 1.5 hour while stirring, followed by washing eight times with 83 ml of heptane at 40 °C, whereby a solid catalyst component having a Ti content of 2.5 % by weight and an Al content of 0.8 % by weight was obtained.

Preparation of Catalyst and Polymerization of Propylene:

Using the above-prepared solid catalyst component, propylene was polymerized in the same manner as in Example 15, except that cyclohexyl methyl dimethoxy silane was replaced with cyclohexyl cyclopentyl dimethoxy silane, and the results are shown in Table 8.

COMPARATIVE EXAMPLE 9

A solid catalyst component was prepared and the polymerization of propylene was carried out in the same manner as in Example 16, except that aluminium trichloride and dimethyl polysiloxane were not used. The results obtained are shown in Table 8.

TABLE 8

	Ex.16	Ex.17	Ex.18	Comp.Ex.9
Polymerization activity (Y)	37,500	43,000	42,000	22,800
RDS*	0.7	0.5	0.5	1.0
Yield of whole crystalline polymer (t-II)	95.6	96.3	96.1	98.5
Density of polymer ( $\rho$ : g/ml)	0.9042	0.9051	0.9038	0.9075
Melt index of polymer (MI: g/10 min)	2.5	1.7	3.0	1.9
Bulk density of polymer (BD: g/ml)	0.41	0.38	0.38	0.40
Content of fine powder (100 $\mu$ or less in diameter)	1.0	1.5	1.5	9.0

(\* in n-heptane)

EXAMPLE 19Preparation of Solid Catalyst Component:

10 g of diethoxymagnesium and 90 ml of toluene were put in a 500-ml round flask equipped with a stirrer, which had been fully purged with nitrogen gas, to prepare a suspension. Then, 20 ml of titanium tetrachloride was added therein, and the temperature was increased to 50 °C while stirring to carry out the reaction. Next, 4.5 ml of di-i-octyl phthalate was added, and the temperature was further increased to 110 °C, and then 4.0 ml of dimethyl polysiloxane having a viscosity of 50 cst at room temperature was added to continue the reaction for 2 hours. After completion of the reaction, a supernatant of the resulting

suspension was removed, and the residue was washed three times with 88 ml of toluene at 75°C. Thereafter, 80 ml of toluene, 1.0 g of aluminium trichloride and 30 ml of titanium tetrachloride were added, and the reaction was carried out at 105°C for 2 hours while stirring, followed by washing eight times with 80 ml of heptane at 40°C, whereby a solid catalyst component having a Ti content of 2.6 % by weight and an Al content of 0.7 % by weight was obtained.

#### Preparation of Catalyst and Polymerization of Propylene:

20 ml of n-heptane was put in a 2000-ml stainless steel autoclave equipped with a stirrer, which had been fully dried with nitrogen gas and then purged with propylene gas. Under the propylene gas atmosphere, 1.32 mmol of triethylaluminium, 0.13 mmol of cyclohexyl methyl dimethoxysilane and 0.0033 mmol (in terms of Ti) of the above-prepared solid catalyst component were added therein, whereby a catalyst was prepared. Thereafter 1,400 ml of liquefied propylene was added therein, and the mixture was stirred at 20°C for 5 minutes. 1500 ml of hydrogen gas was introduced with stirring, and then the temperature in the system was immediately increased to 70°C to carry out the polymerization for 1 hour, whereby 224 g of polypropylene was obtained.

A polymerization activity, which was expressed by a polymer yield per g of the solid catalyst component in a polymerization time of 1 hour, was 37,300 g/g-cat. For evaluation of stereoregularity of the polymer obtained, a content of insoluble polymer after boiling in n-heptane for 6 hours was measured, and it was 95.5%. Further, it was found that the polymer obtained had a density of 0.9052, an average particle size of 400 µm, a bulk density of 0.42 g/ml, and a melt index of 9.4g/10 min. Furthermore, the content of fine powder of 100 micron or less in diameter was 1.5 % by weight.

As described above, when olefins, particularly propylene, are polymerized with the catalyst comprising the solid catalyst component of embodiment 1 of the present invention, the catalytic activity is sufficiently high, so that a residual amount of the catalyst present in the resulting polymer can be suppressed to a very low level, and accordingly, a residual chlorine amount in the resulting polymer can be reduced to an extent that a step of removing such contaminants can be omitted. A density of resulting stereoregular polypropylene can stably be controlled within the range of 0.900 to 0.906 g/ml without changing a process parameter in a polymerization reaction to a large extent, and a polymer suited for a film or sheet formation can readily be produced. Further, in the case where the polymerization is carried out in the presence of the catalyst, the content of fine powders in the resulting polymer can be reduced, and therefore, troubles on a process operation due to such fine powders can be prevented.

Since the RDS rate can be controlled very low, a post-treatment of a polymer produced by the slurry method and a refining process for a polymerization solvent used in the slurry method, can be simplified, resulting in cost reduction such as saving of energy in an operation. Further, the catalyst comprising the solid catalyst component of embodiment 1 of the present invention has a long life with respect to the polymerization activity, enabling steady control of the polymerization process.

The solid catalyst component of embodiment 1 can be prepared by a simple process without any specific additional equipments and exhibits a stable quality with a good reproducibility. Further, the solid catalyst component is advantageous because of its low production cost owing to use of inexpensive materials and a fast settling speed in a washing process as well as a small loss of solid materials during the process.

#### EXAMPLE 20

##### Preparation of Solid Catalyst Component:

80 ml of toluene and 10 g of diethoxymagnesium were put in a flask which had been fully purged with nitrogen gas, and they were mixed to obtain a suspension. The suspension was stirred at room temperature for 10 minutes, and 20 ml of  $TiCl_4$  was then added to the suspension. This liquid mixture was gradually heated and, at the time when the temperature of the mixture had reached 62°C, 1.0 ml of diethyl phthalate was added. The mixture was then heated and, at the time when the temperature thereof had reached 110°C, 3.5 ml of bis(2-ethylhexyl) phthalate was added. The resulting mixture was heated to 112°C and allowed to react at this temperature for 2 hours. After completion of the reaction, the supernatant was removed, and the residual reaction product was washed twice with 100 ml of toluene. Into this flask were introduced 20 ml of  $TiCl_4$  and 80 ml of toluene. This liquid mixture was allowed to react at 100°C for 2 hours. After completion of the reaction, the reaction product was washed 10 times with 100 ml of n-heptane to obtain a solid catalyst component. This solid catalyst component was analyzed to determine ingredients

contained therein. As a result, the ingredient contents were found to be as follows: Ti = 2.46 wt%, Mg = 18.0 wt%, Cl = 62.0 wt%, diethyl phthalate = 6.24 wt%, ethyl 2-ethylhexyl phthalate = 2.90 wt%, bis(2-ethylhexyl) phthalate = 2.50 wt%.

#### 5 Formation of Catalyst and Polymerization

Into a 2-liter autoclave equipped with a stirrer, which had been fully purged with nitrogen gas, were introduced 1.32 mmol of triethylaluminum, 0.13 mmol of dicyclohexyldimethoxysilane, and 0.0066 mmol of the solid catalyst component in terms of the amount of Ti atoms. Thus, a polymerization catalyst was  
 10 formed. Thereafter, 1.8 liters of hydrogen gas and 1.4 liters of liquefied propylene were introduced into the autoclave to conduct polymerization at 70 °C for 30 minutes. The weight of the polymer yielded through the polymerization reaction was 250.5 g. When this polymer was extracted with boiled n-heptane for 6 hours, the weight of the polymer dissolved in the n-heptane was 4.91 g. The catalytic activity was 19,500 g per g of the solid catalyst component used. The yield of the wholly crystalline polymer was 98.3%. The polymer  
 15 yielded had an MI of 3.5 g/10-min, a melting point of 163.0 °C, and a molecular weight distribution of 6.5.

#### EXAMPLE 21

A solid catalyst component was prepared in the same manner as in Example 20, except that diisodecyl  
 20 phthalate was used in place of bis(2-ethylhexyl) phthalate. The solid catalyst component obtained was analyzed to determine ingredients contained therein. As a result, the ingredient contents were found to be as follows: Ti = 2.93 wt%, Mg = 18.3 wt%, Cl = 62.5 wt%, diethyl phthalate = 7.53 wt%, isodecyl ethyl phthalate = 0.51 wt%, isodecyl phthalate = 1.78 wt%.

The formation of a catalyst and polymerization were carried out in the same manner as in Example 20,  
 25 except that the solid catalyst component obtained above was used. As a result, a polymer was obtained in an amount of 200.6 g. The catalytic activity was 18,600 g per g of the solid catalyst component. The yield of wholly crystalline polymer was 98.5%. The polymer yielded had an MI of 2.5 g/10-min, a melting point of 162.5 °C, and a molecular weight distribution of 6.2.

#### 30 EXAMPLE 22

A solid catalyst component was prepared in the same manner as in Example 20, except that 2.5 ml of a mixture of phthalic diesters having alkyl groups with 6 to 12 carbon atoms (trade name "Vinisizer 85,"  
 35 manufactured by Kao Corp., Japan) was used in place of bis(2-ethylhexyl) phthalate. The solid catalyst component obtained was analyzed to determine ingredients contained therein. As a result, the ingredient contents were found to be as follows: Ti = 3.03 wt%, Mg = 18.5 wt%, Cl = 62.8 wt%, diethyl phthalate = 6.23 wt%, ethyl n-octyl phthalate = 0.27 wt%, sum of the other phthalic esters = 3.89 wt%.

The formation of a catalyst and polymerization were carried out in the same manner as in Example 20,  
 40 except that the solid catalyst component obtained above was used. As a result, a polymer was obtained in an amount of 202.3 g. The catalytic activity was 19,400 g per g of the solid catalyst component. The yield of wholly crystalline polymer was 98.2%. The polymer yielded had an MI of 2.4 g/10-min, a melting point of 162.8 °C, and a molecular weight distribution of 7.8.

#### EXAMPLE 23

45

#### Preparation of Solid Catalyst Component:

Into a flask which had been fully purged with nitrogen gas were introduced 80 ml of toluene and 10 g of diethoxymagnesium. The contents were stirred to obtain a suspension. The suspension was stirred at room  
 50 temperature for 10 minutes, and 20 ml of TiCl<sub>4</sub> was then added to the suspension. This liquid mixture was gradually heated and, at the time when the temperature of the mixture had reached 62 °C, 1.0 ml of diethyl phthalate was added. The mixture was then heated and, at the time when the temperature thereof had reached 110 °C, 3.5 ml of bis(2-ethylhexyl) phthalate was added. The resulting mixture was heated and, at the time when the temperature thereof had reached 112 °C, 1.0 ml of diisodecyl adipate was added. This  
 55 mixture was then allowed to react at 112 °C for 2 hours. After completion of the reaction, the supernatant was removed, and the residual reaction product was washed twice with 100 ml of toluene. Into this flask were introduced 20 ml of TiCl<sub>4</sub> and 80 ml of toluene. This liquid mixture was allowed to react at 100 °C for 2 hours. After completion of the reaction, the reaction product was washed 10 times with 100 ml of n-

heptane to obtain a solid catalyst component. This solid catalyst component was analyzed to determine ingredients contained therein. As a result, the ingredient contents were found to be as follows: Ti = 2.70 wt%, Mg = 17.9 wt%, Cl = 61.8 wt%, diethyl phthalate = 6.50 wt%, ethyl 2-ethylhexyl phthalate = 2.80 wt%, bis(2-ethylhexyl) phthalate = 2.60 wt%.

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#### Formation of Catalyst and Polymerization:

An experiment was carried out in the same manner as in Example 20, except that the solid catalyst component obtained above was used. As a result, a polymer was obtained in an amount of 182.5 g. The catalytic activity was 17,500 g per g of the solid catalyst component. The yield of wholly crystalline polymer was 98.1%. The polymer yielded had an MI of 6.5 g/10-min, a melting point of 162.6°C, and a molecular weight distribution of 6.9.

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#### EXAMPLE 24

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An experiment was carried out in the same manner as in Example 20, except that cyclohexylmethyl-dimethoxysilane was used in place of the dicyclohexyldimethoxysilane used in catalyst formation. As a result, a polymer was obtained in an amount of 300.6 g. The catalytic activity was 23,400 g per g of the solid catalyst component. The yield of wholly crystalline polymer was 98.5%. The polymer yielded had an MI of 3.0 g/10-min, a melting point of 162.5°C, and a molecular weight distribution of 6.0.

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#### COMPARATIVE EXAMPLE 10

##### Preparation of Solid Catalyst Component:

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Into a flask the inside atmosphere of which had been sufficiently replaced with nitrogen were introduced 80 ml of toluene and 10 g of diethoxymagnesium. The contents were stirred to obtain a suspension. The suspension was stirred at room temperature for 10 minutes, and 20 ml of  $\text{TiCl}_4$  was then added to the suspension. This liquid mixture was gradually heated and, at the time when the temperature of the mixture had reached 90°C, 2.7 ml of dibutyl phthalate was added. The resulting mixture was heated and continuously stirred at 115°C for 2 hours to allow a reaction to proceed. After completion of the reaction, the reaction product was washed twice with 100 ml of toluene heated at 90°C. To the washed reaction product were added 20 ml of  $\text{TiCl}_4$  and 80 ml of toluene. This mixture was heated to 115°C and allowed to react with stirring at this temperature for 2 hours. After completion of the reaction, the reaction product was washed 10 times with 100 ml of n-heptane warmed at 40°C to obtain a solid catalyst component. This solid catalyst component was analyzed to determine ingredients contained therein. As a result, the ingredient contents were found to be as follows: Ti = 2.91 wt%, Mg = 19.5 wt%, Cl = 63.0 wt%, diethyl phthalate = 1.45 wt%, butyl ethyl phthalate = 2.06 wt%, dibutyl phthalate = 7.28 wt%.

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##### Formation of Catalyst and Polymerization:

The formation of a polymerization catalyst and polymerization were carried out in the same manner as in Example 20, except that the solid catalyst component obtained above was used and cyclohexylmethyl-dimethoxysilane was used in place of dicyclohexyldimethoxysilane. As a result, a polymer was obtained in an amount of 322.7 g. The catalytic activity was 29,700 g per g of the solid catalyst component. The yield of wholly crystalline polymer was 98.4%. The polymer yielded had an MI of 5.0 g/10-min, a melting point of 161.9°C, and a molecular weight distribution of 5.4.

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#### COMPARATIVE EXAMPLE 11

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##### Preparation of Solid Catalyst Component:

Into a flask which had been fully purged with nitrogen gas were introduced 60 ml of toluene and 10 g of diethoxymagnesium. The contents were stirred to obtain a suspension. The suspension was stirred at room temperature for 10 minutes, and 40 ml of  $\text{TiCl}_4$  was then added to the suspension. This liquid mixture was gradually heated and, at the time when the temperature of the mixture had reached 90°C, 2.0 ml of phthaloyl dichloride was added. The resulting mixture was heated to 115°C and continuously stirred at this temperature for 2 hours to allow a reaction to proceed. After completion of the reaction, the reaction product

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was washed twice with 200 ml of toluene heated at 90°C. To the washed reaction product were added 60 ml of toluene and 40 ml of  $\text{TiCl}_4$ . This mixture was heated to 115°C and allowed to react with stirring at this temperature for 2 hours. After completion of the reaction, the reaction product was washed 10 times with 200 ml of n-heptane warmed at 40°C to obtain a solid catalyst component. This solid catalyst component was analyzed to determine ingredients contained therein. As a result, the ingredient contents were found to be as follows: Ti = 3.59 wt%, Mg = 18.6 wt%, Cl = 61.9 wt%, diethyl phthalate = 8.50 wt%.

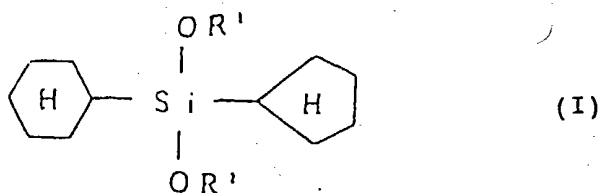
#### Formation of Catalyst and Polymerization:

The formation of a polymerization catalyst and polymerization were carried out in the same manner as in Example 20, except that the solid catalyst component obtained above was used and cyclohexylmethyldimethoxysilane was used in place of dicyclohexyldimethoxysilane. As a result, a polymer was obtained in an amount of 237.3 g. The catalytic activity was 27,000 g per g of the solid catalyst component. The yield of wholly crystalline polymer was 97.8%. The polymer yielded had an MI of 7.1 g/10-min, a melting point of 161.5°C, and a molecular weight distribution of 5.2.

When an olefin is polymerized in the presence of a catalyst comprising the solid catalyst component of embodiment 2, the polymer yielded has extremely high crystallinity. This is supported by the fact that the polymers obtained in the Examples had a melting point higher by about 1°C than those of conventionally known polymers. The catalyst also yielded a stereoregular polymer in high yield and mostly attained a high catalytic activity. Furthermore, a polymer having a broad molecular weight distribution is obtained in high yield. These functions bring about the effect of significantly improving not only the mechanical strength of polypropylene, such as rigidity, i.e., flexural modulus, and impact resistance, but also the physical strength thereof, such as heat resistance. Since the polypropylene thus produced has a broad molecular weight distribution, it also has greatly improved moldability. Thus, the present invention is capable of advantageously providing polypropylene usable in a wide range of applications.

#### Claims

1. A catalyst for polymerization of olefins comprising (A) a solid catalyst component essentially containing magnesium, titanium, an electron donor compound, and a halogen which is prepared by bringing a magnesium compound, a titanium halide compound, and an electron donor compound into contact, (B) an organoaluminum compound, and (C) an organosilicon compound represented by formula (I):



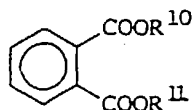
wherein  $\text{R}^1$  and  $\text{R}^2$ , which may be the same or different, each represents an alkyl group having from 1 to 3 carbon atoms; and the cyclohexyl group and the cyclopentyl group may be substituted.

2. The catalyst as in claim 1, wherein said solid catalyst component (A) is prepared using substances (a) to (d) and optionally substance (e):
  - (a) a dialkoxymagnesium represented by  $\text{Mg}(\text{OR}^6)_2$ , wherein  $\text{R}^6$  represents a straight chain or branched chain alkyl group or an aryl group;
  - (b) an aluminium compound represented by  $\text{Al}(\text{OR}^7)_m\text{X}^{2-3-m}$ , wherein  $\text{R}^7$  represents a straight chain or branched chain alkyl group;  $\text{X}^2$  represents a halogen atom; and  $m$  is 0 or integer of 1 to 3;
  - (c) titanium tetrachloride,
  - (d) a diester of aromatic dicarboxylic acid; and
  - (e) a dimethyl polysiloxane.
3. The catalyst as in claim 1, wherein said solid catalyst component is prepared by contacting compounds (a') to (d'):

(a') a magnesium compound represented by  $Mg(OR^8)_{2-p}X^3_p$ , wherein each  $R^8$  independently represents an alkyl group having 1 to 4 carbon atoms or a phenyl group,  $X^3$  represents chlorine or bromine, and  $p$  represents 0 or 1;

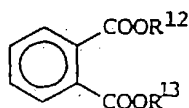
(b') a titanium compound represented by  $Ti(OR^9)_qX^4_{4-q}$  wherein  $R^9$  represents an alkyl group having 1 to 4 carbon atoms,  $X^4$  represents a halogen element, and  $q$  is 0, 1, 2 or 3;

(c') at least one phthalic diester represented by



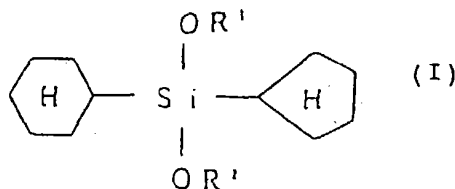
wherein  $R^{10}$  and  $R^{11}$  may be the same or different and each represents an alkyl group having 1 to 4 carbon atoms; and

(d') at least one phthalic diester represented by



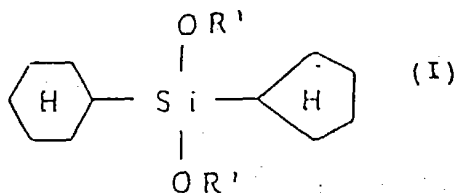
wherein  $R^{12}$  and  $R^{13}$  may be the same or different and each represents an alkyl group having 4 to 12 carbon atoms, provided that the total number of carbon atoms of  $R^{12}$  and  $R^{13}$  is from 10 to 20.

4. A Ziegler-Natta catalyst for olefin polymerization, containing as an effective electron donor ingredient an organosilicon compound represented by formula (I)



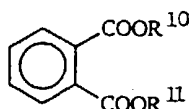
wherein  $R^1$  and  $R^2$ , which may be the same or different, each represents an alkyl group having 1 to 3 carbon atoms, and the cyclohexyl group and the cyclopentyl group may be substituted.

5. An organosilicon compound represented by formula (I)



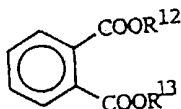
wherein  $R^1$  and  $R^2$ , which may be the same or different, each represents an alkyl group having 1 to 3 carbon atoms, and the cyclohexyl group and the cyclopentyl group may be substituted.

6. The organosilicon compound as claimed in claim 5, which is cyclohexylcyclopentylmethoxysilane, cyclohexylcyclopentyl-diethoxysilane, cyclohexylcyclopentyl-di-n-propoxysilane, or cyclohexylcyclopentyl-diisopropoxysilane.
7. A solid catalyst component for polymerizing olefins, which is prepared using substances (a) to (d) and optionally substance (e):
- (a) a dialkoxymagnesium represented by  $\text{Mg}(\text{OR}^6)_2$ , wherein  $\text{R}^6$  represents a straight chain or branched chain alkyl group or an aryl group;
  - (b) an aluminium compound represented by  $\text{Al}(\text{OR}^7)_m \text{X}^{2-3-m}$ , wherein  $\text{R}^7$  represents a straight chain or branched chain alkyl group;  $\text{X}^2$  represents a halogen atom; and  $n$  is 0 or integer of 1 to 3;
  - (c) titanium tetrachloride,
  - (d) a diester of aromatic dicarboxylic acid; and
  - (e) a dimethyl polysiloxane.
8. A solid catalyst component for polymerizing olefins, which is prepared by contacting compounds (a') to (d'):
- (a') a magnesium compound represented by  $\text{Mg}(\text{OR}^8)_2 \text{X}^3_p$ , wherein each  $\text{R}^8$  independently represents an alkyl group having 1 to 4 carbon atoms or a phenyl group,  $\text{X}^3$  represents chlorine or bromine, and  $p$  represents 0 or 1;
  - (b') a titanium compound represented by  $\text{Ti}(\text{OR}^9)_q \text{X}^4_{4-q}$ , wherein  $\text{R}^9$  represents an alkyl group having 1 to 4 carbon atoms,  $\text{X}^4$  represents a halogen element, and  $q$  is 0, 1, 2 or 3;
  - (c') at least one phthalic diester represented by



wherein  $\text{R}^{10}$  and  $\text{R}^{11}$  may be the same or different and each represents an alkyl group having 1 to 4 carbon atoms; and

(d') at least one phthalic diester represented by



wherein  $\text{R}^{12}$  and  $\text{R}^{13}$  may be the same or different and each represents an alkyl group having 4 to 12 carbon atoms, provided that the total number of carbon atoms of  $\text{R}^{12}$  and  $\text{R}^{13}$  is from 10 to 20.

9. A process for polymerizing an olefin, comprising homo- or copolymerizing an olefin in the presence of the catalyst as claimed in any of claims 1 to 3.
10. A process for polymerizing an olefin, comprising homo- or copolymerizing an olefin in the presence of a catalyst containing the solid catalyst component as claimed in any of claim 7 and 8.

FIG. 1

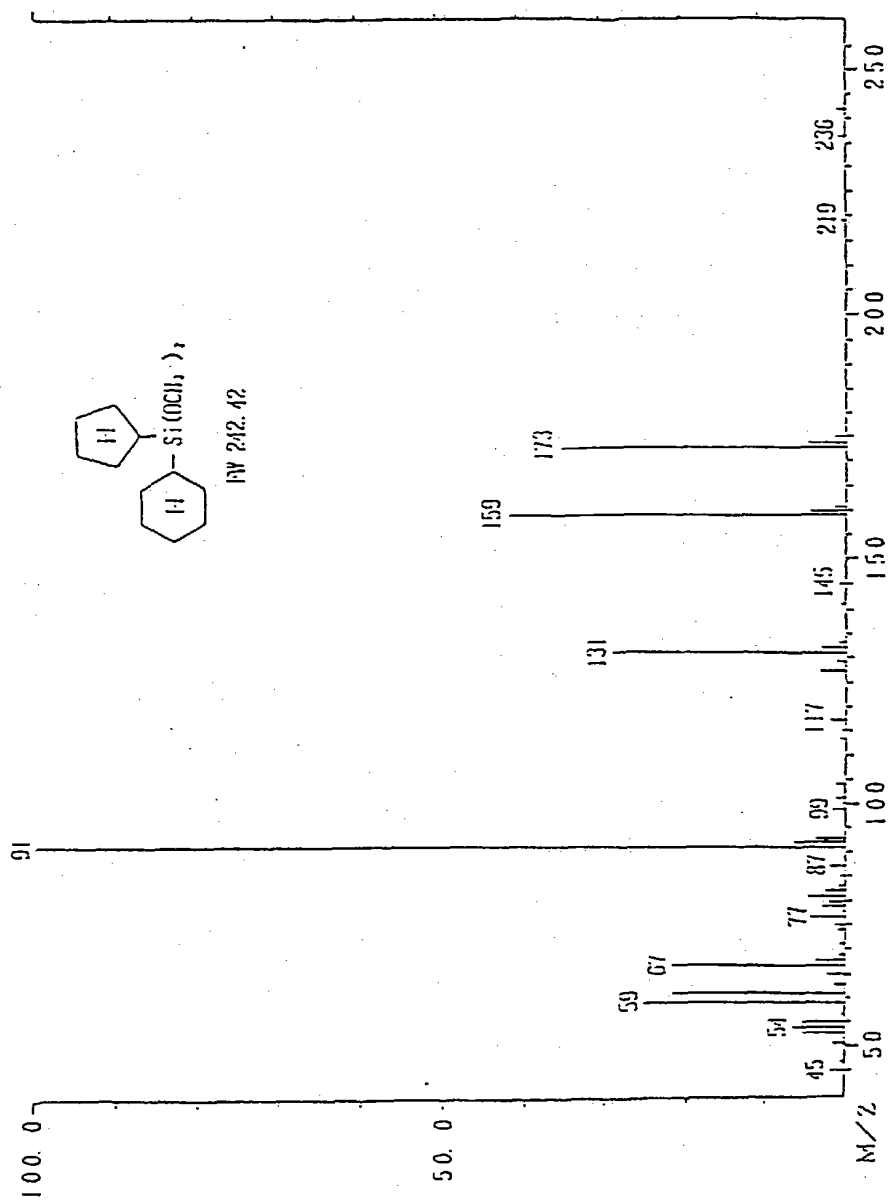


FIG. 2

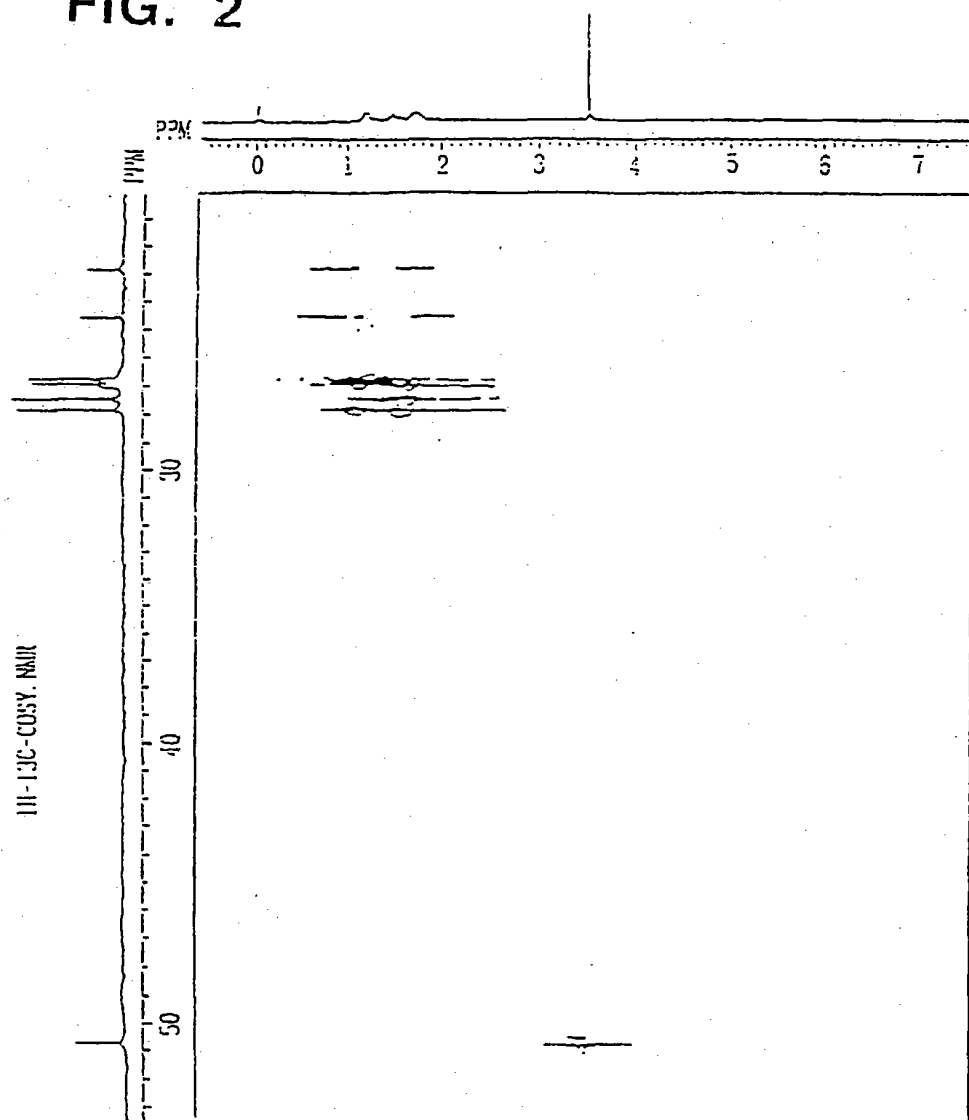


FIG. 3

